JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 99, NO. D12, PAGES 25,553–25,585, DECEMBER 20, 1994

Trends in the vertical distribution of ozone: An analysis of ozonesonde data

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Abstract. I present an analysis of trends in ozone since about 1970 and discuss the quality of the ozonesonde data and inconsistencies among data records. In the troposphere there are significant spatial variations in the trends, with largest increases found over Europe (~2% yr^{-1} throughout the troposphere) and no long-term trend over Canada; there is a small (<1% yr^{-1}) increase over the east coast of the United States in summer. The Japanese stations show increases in ozone only below 5.5 km. The trends in tropospheric ozone are discussed in the context of trends in emissions of nitrogen oxides from surface sources and from aircraft. Trends in surface emissions of NO_x have been similar in the United States. and Western Europe, while the trend in ozone has been larger over Europe; the cause of the large increase in ozone over Europe is unclear. The increase over Europe has leveled off in recent years, and there is no increase since 1980 over the eastern United States. The lack of an ozone increase in the last decade over these two regions during a period of rapid growth of aircraft traffic argues against a significant influence from emissions of NO_x from aircraft. The large interannual variability in ozone in the upper troposphere is similar to that in the lower stratosphere. Any short-term trend in ozone near the tropopause could be caused simply by dynamical factors. Stratospheric ozone decreases are found from about 24 km to near the tropopause. Ozone losses below 17 km appear to be responsible for the 20% difference between trends in column ozone derived from the Stratospheric Aerosol and Gas Experiment (SAGE) and the total ozone mapping spectrometer (TOMS). Ozone changes in the troposphere make an important contribution to the column ozone change for some stations. The stratospheric decreases are larger in winter than in summer over Europe and the midlatitude stations of North America; they are larger in summer than in winter over the high latitude (>53°N) stations of North America. These seasonal losses are consistent with the patterns reported by Stolarski et al. (1992) using TOMS data. Losses are found year round over Syowa, Antarctica, although they are largest in spring.

1. Introduction

Ozone plays an important role in the energy budget of the atmosphere, since it absorbs both solar and infrared radiation [*Ramanathan et al.*, 1976; *Ramanathan and Dickenson*, 1979; *Fishman et al.*, 1979a; *Wang et al.*, 1980]. Decreases in ozone in the lower stratosphere cause decreases in surface temperature, while increases in ozone in the upper troposphere cause increases in surface temperature; the resultant effect on surface temperature is critically dependent on the vertical distribution of the trend in ozone [*Lacis et al.*, 1990; *Schwarzkopf and Ramaswamy*, 1993; *Wang et al., 1993]*.

International assessments of trends in the total column of ozone and in stratospheric ozone have relied primarily on ground-based measurements using Dobson or Brewer spectrophotometers, and on satellite borne instruments, the total ozone mapping spectrometer (TOMS) and the Stratospheric Aerosol and Gas Experiment (SAGE) [e.g., *World Meteorological Organization* (hereinafter WMO), 1990a,b, 1992; *Bojkov et al.*, 1990; *Stolarski et al.*, 1992; *McCormick et al.*, 1992]. Much less attention has been given in these assessments to measurements of the vertical distribution of ozone using ozonesondes. There are a number of difficulties with the ozonesonde data [e.g., *Logan*, 1985; *Tiao et al.*, 1986; *U.S. Department of Energy* (hereinafter DOE), 1993], but they provide the only data on trends in tropospheric ozone above the surface and on trends in the stratosphere below 17 km; they also provide unique information on stratospheric trends below 30 km prior to the satellite measurements that commenced in 1979.

Comprehensive analyses of ozonesonde data obtained prior to 1983 indicated that ozone was increasing in the troposphere and decreasing in the lower stratosphere at many of the northern hemisphere stations [*Angell and Korshover*, 1983; *Angell et al.*, 1985; *Logan*, 1985; *Tiao et al.*, 1986]. Results from more recent studies offer mixed conclusions. *Bojkov* [1988], *London and Liu* [1992], *London* [1993], and *Wang et al.* [1993] argue that tropospheric ozone is increasing at northern midlatitudes; *Oltmans* [1993] shows that tropospheric ozone has been decreasing at the two Canadian polar stations since 1980. An update of *Tiao et al.* [1986] to include data up to 1986 reports decreases in tropospheric

Paper number 94JD02333.
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ozone for three of the four Canadian stations and increases for the European and Japanese stations [*WMO*, 1990b].

I present here a comprehensive trend analysis of ozonesonde data up to the end of 1991; this period was selected to allow comparison with satellite observations. Data from later years are discussed where appropriate. Problems with data quality and inconsistencies among data records are highlighted. Trends in tropospheric ozone are discussed in the context of trends in emissions of NO_x, the limiting species for ozone formation in most of the troposphere [e.g., *Chameides et al.*, 1992]. Trends in stratospheric ozone are compared with results from TOMS and SAGE.

2. Analysis of Ozone Measurements

Ozonesonde data were obtained from the World Ozone Data Center (WODC) for Resolute, Churchill, Edmonton, and Goose Bay in Canada; Wallops Island in the United States; Hohenpeissenberg, Berlin, and Lindenberg in Germany; Legionowo in Poland; Sapporo, Tateno, and Kagoshima in Japan; Aspendale/Laverton in Australia; and Syowa, Antartica. The data were current as of August 1994. Surface data for Hohenpeissenberg were obtained also from WODC. J. Staehelin provided a revised data set for Payerne, Switzerland, in November 1993. Recent data for Boulder, in the United States, were provided by S. Oltmans, and data for 1963-1966 were taken from *Dutsch et al.*, [1970]. Data for Natal, Brazil, were obtained from A. Torres and V. Kirchhoff.

2.1. Data Quality

Two general types of ozonesonde are in use today, variations on the Brewer Mast (BM) bubbler [*Brewer and Milford*, 1960] and on the electrochemical concentration cell (ECC) [*Komhyr*, 1969]; all are based on the reaction of KI with ozone. Difficulties associated with these instruments and with interpretation of the data are discussed by *Logan* [1985], *Tiao et al.* [1986], *U.S. DOE* [1993], and references therein. Sulphur dioxide interferes negatively with the electrochemical technique on a molar basis, while NO_2 gives a small positive interference of a few percent [*Schenkel and Broder*, 1982]. A decrease in ambient SO_2 would mimic an increase in ozone [*Logan*, 1985]. Emissions of SO_2 decreased by 25% in the United States from 1970 to 1990 [*EPA*, 1992a], and by 74% in Japan from 1970 to 1980 [*Organization for European Cooperation and Development* (hereinafter OECD), 1993]. In the former West Germany, emissions of SO_2 decreased by 15% from 1970 to 1980, and by 70% from 1980 to 1990 [*OECD*, 1993].

Brewer Mast sondes underestimate ozone, in part because of the strength of the KI solution [*Powell and Simmons*, 1969]. The vertical profile of ozone is integrated, allowing for the amount of ozone above the altitude reached by the sonde, and scaled to a concurrent measurement of the ozone column. All sonde measurements except those from Wallops Island after May 1982 and those from Natal had been scaled to the ozone column. The scaling, or correction factor (CF), is typically about 1.25 for BM sondes and about 1.0 for ECC soundings (see Table 1). There are

Table 1a. Correction Factors for Brewer Mast Stations

Station	Dates	Number	Fraction With CF	Mean CF	CF = 0.9-1.2		CF = 0.9-1.35	
					Fraction	Mean CF	Fraction	Mean CF
Resolute[a]	1966-1979	608	0.62	1.23	0.48	1.11	0.81	1.17
Churchill	1973-1979	286	1.00	1.27	0.37	1.10	0.64	1.17
Edmonton	1972-1979	357	1.00	1.26	0.47	1.10	0.75	1.17
Goose Bay	1970-1980	475	1.00	1.28	0.34	1.10	0.72	1.20
Berlin	1966-1973	357	1.00	1.33	0.31	1.12	0.68	1.20
Lindenberg[b]	1975-1991	1147	1.00	1.24	0.47	1.08	0.71	1.14
Legionowo[c]	1979-1991	434	1.00	1.41	0.22	1.08	0.46	1.18
Hohenpeissenberg[d]	1970-1993	2474	1.00	1.09	0.91	1.08	0.99	1.09
Payerne[e]	1970-1991	3117	0.99	1.23	0.49	1.11	0.82	1.17
Aspendale/Laverton[f]	1965-1990	592	0.99	1.23	0.44	1.10	0.79	1.18

The fourth column gives the fraction of soundings with a correction factor (CF), the fifth column gives the mean of all CFs, the sixth column gives the fraction of CFs in the range 0.9-1.2, the seventh column gives the mean of these CFs, and the seventh and eighth columns give the same for the range 0.9-1.35.

[a] Column measurements cannot be made in winter at Resolute, and a mean correction factor of 1.186 was given for these soundings.

[b] A Brewer type of sonde developed and manufactured in the former East Germany was used at Lindenberg [*Feister et al.*, 1985] until March 1992, when ECC sondes were adopted. The data are scaled to column measurements made at Potsdam.

[c] The station used East German sondes until June 1993.

[d] Data are available for 1967-1969, but there were no correction factors given until late 1969. Dobson measurements were not started at Hohenpeissenberg until 1968.

[e] Data from Payerne are scaled to column measurements made at Arosa, 200 km distant.

[f] The time series for Aspendale and Laverton have been combined (the station was moved in 1982), and are treated in all analyses as one station. A few test ECC sondes were flown in late 1990.

Table 1b. Correction Factors for ECC Stations

Station	Dates	Number	Fraction With CF	Mean CF	Fraction With CF = 1.0	CF = 0.8-1.2	
						Fraction	Mean CF
Resolute	1980-1993	569	1.00	1.01	0.51	0.96	1.00
Churchill	1980-1993	538	1.00	1.00	0.36	0.95	1.00
Edmonton	1980-1993	534	1.00	1.02	0.21	0.96	1.00
Goose Bay	1980-1993	582	1.00	1.02	0.38	0.94	1.01
Wallops Island*	1970-1981	468	0.78	1.00	0.01	0.94	0.99
Sapporo	1970-1993	360	0.98	1.02	0.00	0.88	0.99
Tateno	1970-1993	507	0.99	1.01	0.00	0.93	1.00
Kagoshima	1970-1993	327	0.99	0.98	0.00	0.89	0.98
Natal	1978-1991	253	0.0				
Syowa	1966-1992	413	0.83	1.05	0.06	0.84	1.01

The fourth column gives the fraction of soundings with a correction factor (CF), the fifth column gives the mean of all CFs, the sixth column gives the fraction of CFs with the value 1.000 (used when there was no column measurement for the Canadian stations), the seventh column gives fraction of CFs in the range 0.8-1.2, and the eighth column gives the mean of these CFs.

* Wallops Island stopped giving correction factors in May 1982; data are available from WODC up to early 1993. A. Torres provided 2 years of data that were missing from the WODC archive for Wallops Island.

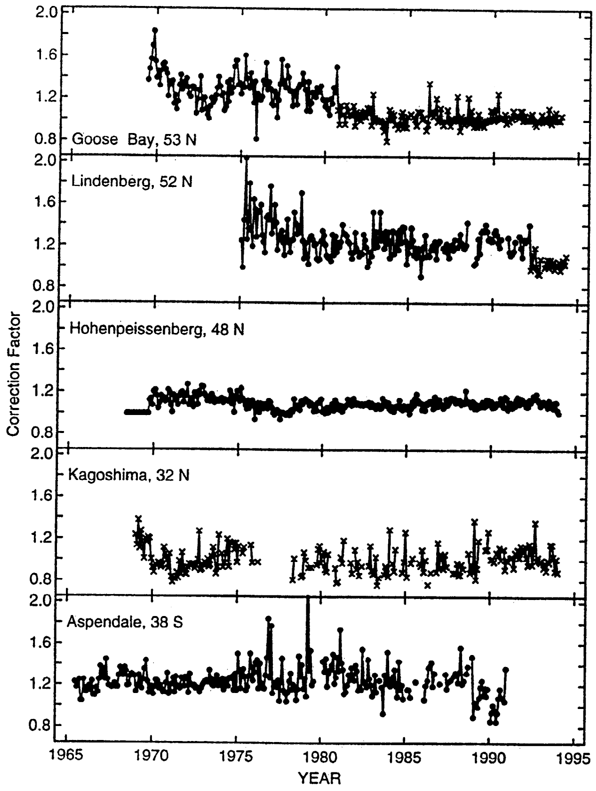


Figure 1. Correction factors for selected ozonesonde stations, shown as monthly mean values. All correction factors were included. Brewer Mast data are shown as solid circles, ECC data as crosses.

difficulties associated with this practice: the use of a scaling factor may distort the shape of the vertical profile, and any errors in the column measurements are carried over into the sonde results. *De Muer* [1985] argued that the correction factor should be height dependent (largest in the troposphere), while *Hilsenrath et al.* [1986] suggested a constant additive correction for BM sondes; these approaches have not been implemented for the data analyzed here.

The correction factors are used as a quality check; they depend primarily on the reliability of the measurement of stratospheric ozone. ECC soundings were included in the analysis if the correction factor was in the range 0.8-1.2, about 90% of the soundings at most stations (see Table 1). For BM stations, soundings were included if the correction factor was in the range 0.9-1.35, 45-80% of the soundings, depending on the station. A narrower range, 0.9-1.2, was used for Hohenpeissenberg, as this includes >90% of the soundings; this range would exclude 50-80% of the soundings at the other BM stations, with a bias toward loss of summer measurements. Figure 1 shows time series of monthly mean correction factors for selected stations.

There are statistically significant trends in the correction factors for some stations, as shown in Table 2. The trend is largest for Lindenberg and Aspendale, for the period after 1980. The correction factor for BM sondes is sensitive to preflight preparation procedures [*Staehelin and Schmid*, 1991], and a change in procedure may be responsible for the shift in values at Lindenberg in 1979 and at Aspendale in early 1980. Errors in the measurement of the ozone column provide another possible cause of trends in the correction factors.

Bojkov et al. [1990] revised the ground-based ozone column data for the last WMO assessment [*WMO*, 1992]. The revisions to monthly mean data were based on comparisons with TOMS for

Table 2. Trend in Correction Factors

Station	CF Selected	Year	Trend, % yr^{-1}
Edmonton	0.9-1.35	1972-1979	(0.60 ± 0.66)
	0.8-1.2	1980-1991	-0.32 ± 0.24
Goose Bay	0.9-1.35	1970-1979	(-0.29 ± 0.41)
Legionowo	0.9-1.35	1980-1991	0.57 ± 0.47
Lindenberg	0.9-1.35	1980-1991	0.76 ± 0.31
Hohenpeissenberg	0.9-1.2	1980-1991	0.28 ± 0.12
Payerne	0.9-1.35	1970-1991	-0.44 ± 0.08
	0.9-1.35	1980-1991	-0.65 ± 0.23
Sapporo	0.8-1.2	1980-1991	(-0.16 ± 0.54)
Tateno	0.8-1.2	1980-1991	(-0.20 ± 0.34)
Kagoshima	0.8-1.2	1980-1991	0.57 ± 0.54
Aspendale	0.9-1.35	1980-1990	-0.82 ± 0.49

Trends were calculated for all stations given in Table 1 for 1970-1991, for 1980-1991, and for the period of Brewer Mast soundings at the Canadian stations. Correction factors were included in the trend calculation only if they fell in the range given in column 2. Trends larger than 0.15% yr^{-1} are shown. The trend is given with twice the standard error. Data from the months of June-August were omitted for the Japanese stations.

data after 1979 [*Bojkov et al.*, 1988], on correlations of the ozone column and 100-mbar temperatures, and on comparisons between neighboring stations. The revisions have not been verified by the individual stations, for Canada, for example (J. Kerr, personal communication, 1993), and have not usually been applied to the sonde data at WODC.

Labow and McPeters [1993] recently presented a comparison of TOMS and ground-based measurements of the ozone column. There were discontinuities in the time series of the difference plots, (TOMS minus ground-based)/TOMS, of as much as 4% for Potsdam (used to scale the Lindenberg sondes), Churchill, and Kagoshima; several of the other stations showed small discontinuities (1-2%). These discontinuities provide further evidence of potential errors in the column data used to scale the sonde data.

Figure 2 shows a comparison of monthly mean values formed from the column data reported for each sounding and the "provisionally revised" column data from *WMO* [1992]. There are periods when there is an obvious offset between the two estimates of the ozone column. Table 3 gives the ratio of the two estimates, while Table 4 gives the trend in the difference between the two. These results are used below in a preliminary assessment of the effects of errors in the column data on trends derived for the vertical distribution of ozone.

The difference between the revised column data and the column data associated with the sondes for Goose Bay (prior to 1980), Hohenpeissenberg, and Kagoshima may be responsible in part for the trend in the correction factors shown in Table 2. The trend in the difference between the two measures of the ozone column (Table 4) is consistent with the trend in the correction factor, if the sondes measure ozone on a reproducible basis. This effect cannot be responsible for the trend in the correction factor at Edmonton, as the sign of the trend is wrong. At Edmonton the time of the soundings changed from early morning to late afternoon in late 1985; the ozone column is about 4 DU (1 DU = 2.69 x 10^{16} molecules cm^{-2}) higher in the afternoon (J. Kerr, personal communication, 1993). The small increase in the column measurement used to scale the soundings may be responsible in part for the small shift in 1985 (Figure 2). The effect of trends in the correction factor or errors in the column data used to determine the correction factors is assessed in section 3 by determining trends with the correction factors removed from the data.

The change from Brewer Mast to ECC sondes in Canada around 1980 caused a shift in values recorded for tropospheric ozone [e.g., *Logan*, 1985]. *Tiao et al.* [1986], using a statistical model, estimated that the shift was 10-20% for Resolute, ~20% for Goose Bay, 20-30% for Churchill, and 20-40% for Edmonton. Intercomparisons in 1970, 1978, and 1984 had shown that ECC sondes measured more ozone in the midtroposphere than BM sondes by about 15 to 20% (with a range of 7-38%); in the stratosphere, BM sondes gave more ozone than ECC sondes, usually by less than 5% [*Attmanspacher and Dutsch*, 1970, 1981; *Hilsenrath et al.*, 1986]. An intercomparison in 1991 gave opposite results [*WMO*, 1991]. Brewer Mast sondes from Hohenpeissenberg gave tropospheric values that were 10-25% higher than ECC sondes provided by Canada and the National Oceanic and Atmospheric Administration (NOAA). In the stratosphere, the BM sondes gave ozone values 2-4% higher than the ECC sondes below 24 km (30 mbar), but gave values 2-4% lower than ECC sondes for 24-30 km (30-10 mbar). These results raise the question of whether there has been an increase in the sensitivity of BM sondes to tropospheric ozone, or a decrease in sensitivity of ECC sondes, with unfortunate implications in either case for the derivation of reliable trends. The ECC sondes from three different groups were self-consistent, and this finding suggests that the BM sondes may

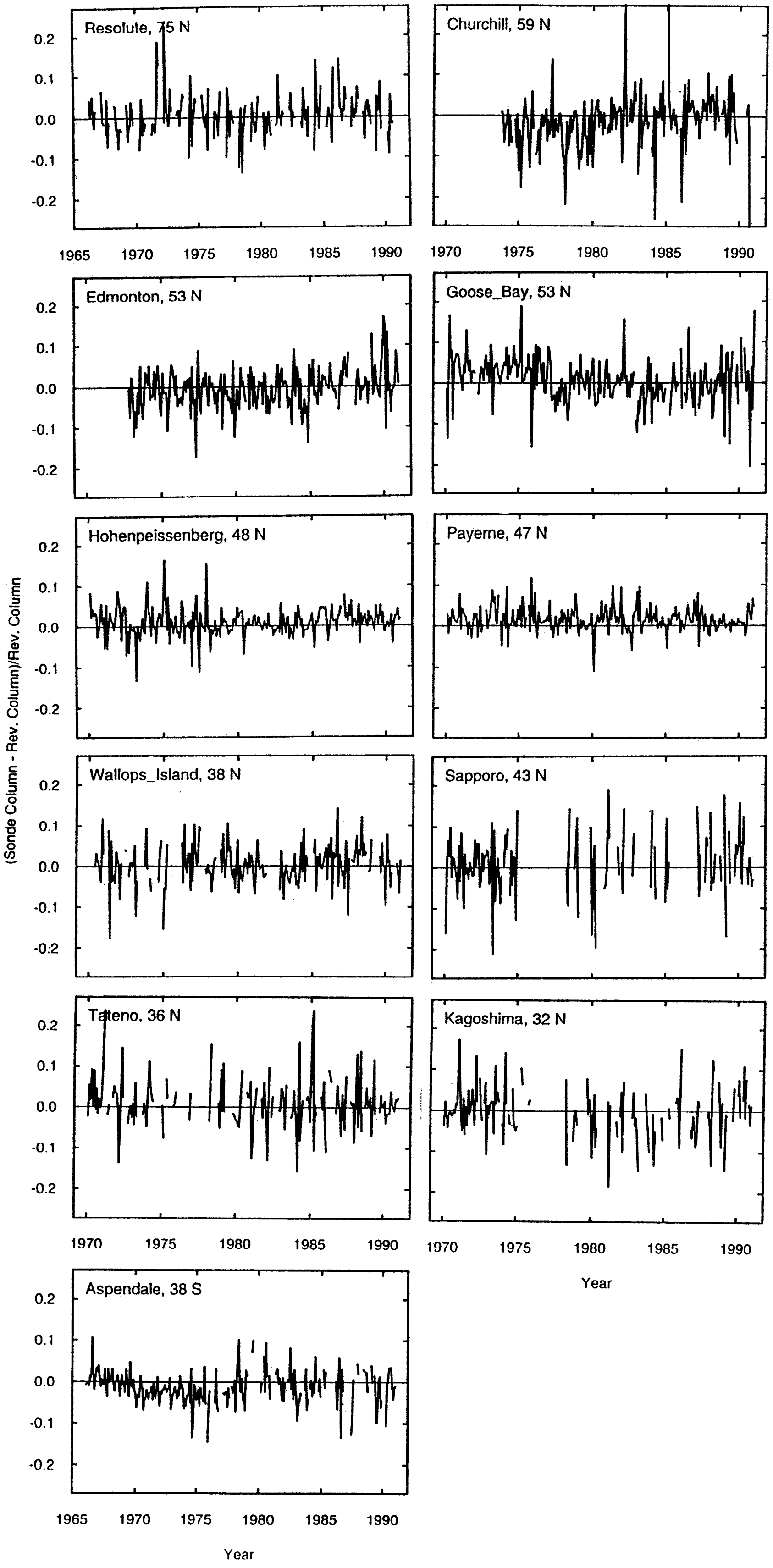


Figure 2. Ratio of monthly mean ozone columns formed from the ground-based column measurements associated with each ozone sounding, and the provisionally revised ozone columns used in the WMO assessment [*WMO*, 1992]. There is scatter in the plots because the soundings are made 2-12 times a month, while the column is measured daily.

Table 3. Ratio of Sonde Column to Corrected Column

Station	Period	Sonde Column / Revised Column
Churchill	October 1973 to December 1979	0.963
Edmonton	October 1972 to December 1984	0.985
Goose Bay	June 1969 to February 1977	1.037
	March 1977 to May 1978	0.961
	January 1980 to December 1984	0.974
Hohenpeissenberg	May 1985 to December 1991	1.018
Kagoshima	January 1978 to March 1989	0.981
	April 1989 to March 1991	1.018
Aspendale	January 1965 to December 1966	1.021
	December 1969 to December 1977	0.971

The table gives the ratio of monthly mean values of the ground-based ozone columns associated with each ozone sounding to the revised column monthly means used in the 1991 ozone assessment [*WMO*, 1992] for the period shown (see Figure 2). The results are given as a trimmed mean, such that the lowest and highest 25% of the ratios were excluded. Ratios for stations not shown are in the range 0.99-1.01. The 1991 ozone assessment did not provide revised column data for Lindenberg (Potsdam), Legionowo, Natal, or Syowa.

have changed. The results of the earlier intercomparisons are consistent with the observed change in ozone values around 1980 in Canada and are used below to assess long-term changes in tropospheric ozone. The Canadian data are treated as a self-consistent time series only in the stratosphere, with the caution that there may be a small offset (<5%) around 1980.

A similar problem arises in merging time series from Berlin, where BM sondes were used from 1966 to 1973, and from Lindenberg, 60 km distant, where Brewer sondes manufactured in the former East Germany (GDR) were used after 1975. Tropospheric values recorded at Lindenberg were about 30% higher than values at Berlin and other European stations in the 1970s [*Logan*, 1985].

Table 4. Trend in Sonde Column minus Corrected Column

Station	CF	Years	Trend, % yr^{-1}
Edmonton	note 1	1973-1990	0.23 ± 0.13
	0.8-1.2	1980-1990	0.41 ± 0.27
Churchill	note 1	1974-1990	0.31 ± 0.22
Goose Bay	note 1	1970-1990	-0.24 ± 0.11
	0.9-1.35	1970-1979	-0.44 ± 0.31
Hohenpeissenberg	0.9-1.2	1980-1990	0.15 ± 0.13
Kagoshima	0.8-1.2	1980-1990	0.56 ± 0.44
Aspendale	0.9-1.35	1970-1990	0.12 ± 0.09

The table gives the trend in the difference between monthly mean ozone columns calculated from column measurements used to scale individual ozone soundings and the revised column measurements used in the 1991 ozone assessment, as a percent of the revised column measurements. Column data from the sonde data were included only if the sonde measurement met the correction factor criteria given in section 2. Trends are shown with twice the standard error and are given only if statistically significant. Trends were calculated for 1965-1990, 1970-1990, and 1980-1990, according to data availability.

Note 1. For the Canadian stations, the correction factor criteria were 0.9-1.35 for Brewer Mast soundings, and 0.8-1.2 for EEC soundings (see Table 1).

The intercomparisons in 1970 and 1978 showed that the GDR sonde gave values 6-30% higher than BM sondes in the troposphere and values within 2-3% in the lower stratosphere. The GDR sondes were replaced with ECC sondes in 1992.

2.2. Data Analysis

Ozone concentrations for each sounding that met the correction factor criteria discussed above for BM and ECC sondes were interpolated to give values at 22 pressure levels from 1000 to 10 mbar, and monthly means were formed. There is a strong seasonal cycle in ozone, and it was removed to form time series of monthly anomalies, y (month, year), that formed the basis for the trend analyses:

$$y(\text{month, year}) = \mu(\text{month, year}) - \bar{\mu}(\text{month}) \qquad (1)$$

Here, $\bar{\mu}$ (month) is the average of all monthly means for a given month over the selected period of the data record.

Monthly means and monthly anomalies are shown for 500 mbar in Figure 3 and for 90 mbar in Figure 4. The change from BM to ECC sondes in Canada, and from BM to GDR to ECC sondes at Berlin/Lindenberg, is indicated by the change in symbol type, and there are discontinuities in the record for 500 mbar but not for 90 mbar in each case. Measurements are made 2-3 times a week at Hohenpeissenberg and Payerne, weekly in Canada and Poland, and once or twice a month in Japan and Australia, with a clear increase in the variance as the frequency declines. There are few summer measurements for most of the record from Japan before 1990. The measurement frequency was reduced from weekly to twice a month in 1985 at Wallops Island, with a significant increase in the variance. Measurements at Syowa were made 10-30 times a year from 1966 to 1972, rarely for the next 10 years, and on a frequent basis since 1986, following the discovery of the Antarctic ozone hole [*Chubachi*, 1985; *Iwasaka and Kondoh*, 1987]. Measurements were made at Natal 7-30 times a year [*Kirchhoff et al.*, 1991]. The time series suggest that weekly sampling is the minimum required to obtain representative monthly means.

Much of the analysis presented here relied on a simple statistical model (model 1), with trends in the monthly anomalies calculated by linear regression. Trends given by season used series of monthly anomalies for the months indicated. Recent trend analyses of the ozone column include the dependence of ozone on the quasi-biennial oscillation (QBO), an oscillation in the equatorial zonal winds in the stratosphere, and on the solar flux [e.g., *Bojkov et al.*, 1990; *WMO*, 1992] to account for known sources of variance in the data. I adopted a similar approach for the sonde data in model 2:

$$y_t = a\,t + b\,\mathrm{QBO}_{t-k} + c\,\mathrm{SOL}_t + N_t \qquad (2)$$

where y_t are the monthly anomalies, t is the time from the start of the period of interest, a is the linear trend, QBO is the 30-mbar zonal wind (in knots) at Singapore lagged k months, SOL is the 10.7-cm solar radio flux, a proxy for solar variability, and N_t is the residual time series. Model 2 was used to ascertain whether the inclusion of QBO and solar flux terms affected the trends and standard errors derived for the sonde data, and was applied to the annual trend and to 4-month seasons, December-March and May-August. Model 2 was fitted for lags k in the range -15 to 25, from 100 to 10 mbar, to determine the optimal lags for the annual and seasonal trends.

Models 1 and 2 may underestimate the error in the ozone trend, because they do not account for temporal correlation in the ozone monthly means [*WMO*, 1990a, Appendix A; *Tiao et al.*, 1990]. This issue is investigated in the appendix. A model including temporal correlation could not be applied uniformly to the sonde data because 20-50% of the monthly means are missing for seven of the stations. The reader is cautioned that the true standard errors may be of the order of 20-60% larger than the standard errors computed with models 1 and 2, as discussed in the appendix. When trends are described as "significant" in the text, they are statistically significant at the 95% confidence level, according to the Student's t test, using model 1 or 2.

3. Results

3.1. The Effect of Uncertainties in Ozone Column Measurements on Trends

Annual trends in ozone versus altitude are shown in Figures 5 and 6 for the periods 1970-1991 and 1980-1991, respectively. Model 1 was used, and all months were weighted equally. These results are used primarily for evaluation of issues relating to data quality and choice of statistical model. Seasonal trends are discussed separately for the troposphere and stratosphere below. The solid lines in Figures 5 and 6 show results obtained by using the sonde data without modification, the long-dashed lines show the effect of adjusting the data to the revised ozone columns by using the ratios in Table 3, and the short-dashed lines show the effect of dividing the data by the correction factor for each sounding to remove the scaling to the ozone column. The effect of the correction factor was not removed for the Canadian stations because of the change in sonde type, except for 1980-1991, or for Payerne, where there were significant shifts in the correction factors [*Staehelin and Schmid*, 1991].

Adjusting the sonde data to the revised columns changes the trends in ozone by less than 0.3% yr^{-1}. The fractional change in the lower stratosphere is large, ~40%, for Hohenpeissenberg (1980-1991) and Churchill (1974-1991), and smaller, 10-20%, for Hohenpeissenberg (1970-1991), Edmonton, Goose Bay, and Aspendale; the change is negligible for Kagoshima for 1970-1991, and large, ~45%, for 1980-1991, but the trend is not significant. The fractional change in the troposphere is less than 20%, except for stations where the trend is not significant.

Removing the correction factor from the sonde data changes the trends by less 0.3% yr^{-1} in the stratosphere and by less than 0.5% yr^{-1} in the troposphere; Lindenberg, Legionowo, and Aspendale are exceptions (Figure 6), where there are large trends in the correction factors after 1980. There is no effect on the standard error of the trends. The fractional change in stratospheric ozone is large, ~40%, for Hohenpeissenberg (1980-1991), and smaller for Edmonton and Goose Bay; it is large for the Japanese stations for 1980-1991, but the trends are not significant. For the troposphere, the fractional change is usually less than 25% when the trends are significant.

Uncertainties in the ozone column measurements affect the magnitude of the trends, but they do not affect the overall pattern of decreases in ozone in the lower stratosphere and increases for some stations in the troposphere and decreases or no trend for others. Lindenberg, Legionowo, and Aspendale are exceptions, where removing the scaling to the ozone column gives trends (after 1980) that are 1-2% yr^{-1} different from those obtained by using the scaled data.

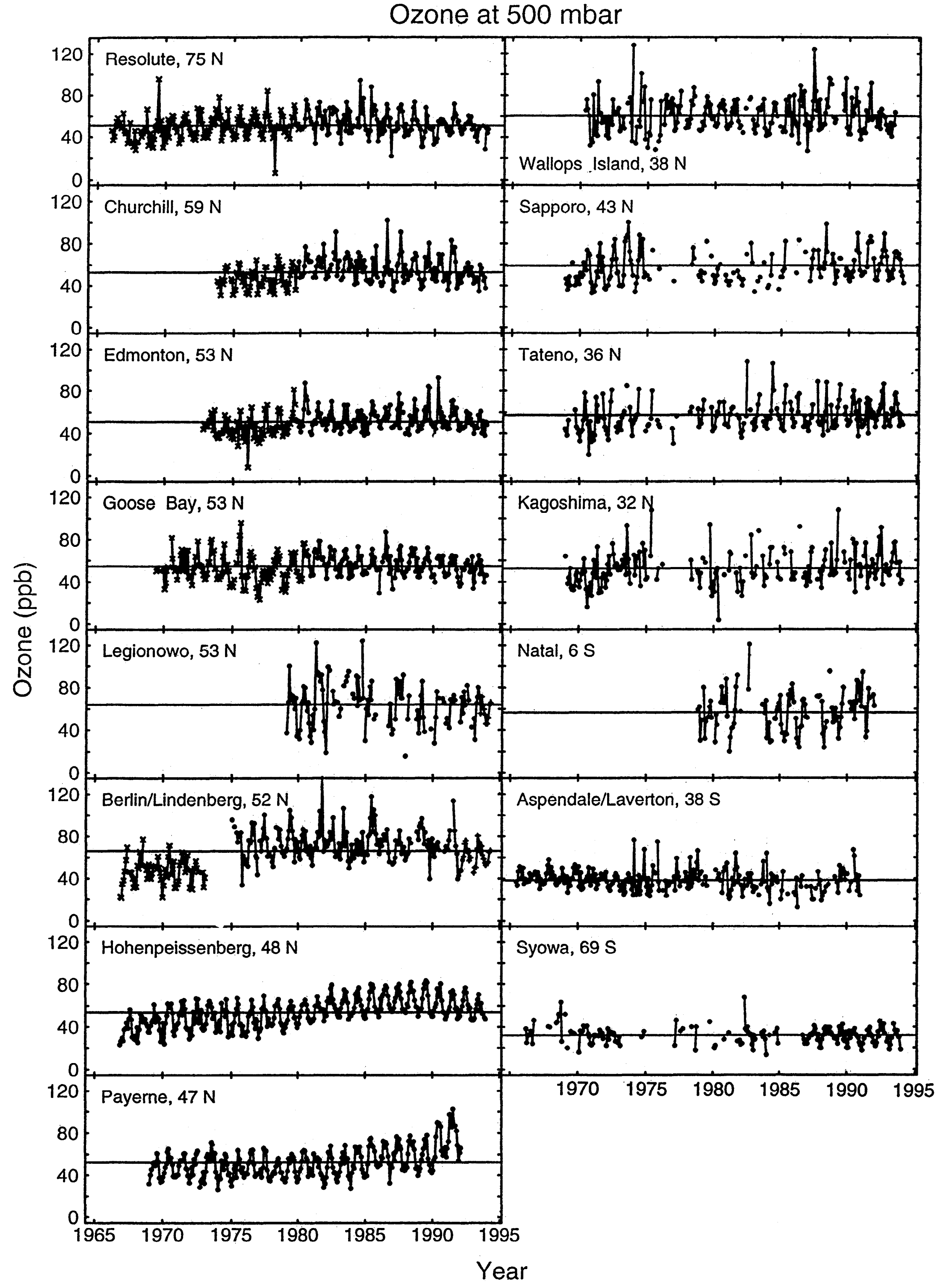


Figure 3a. Monthly mean values of ozone at 500 mbar for each ozonesonde station, given in parts per billion. Soundings were included only if the correction factors met the criteria given in Section 2. The change from BM to ECC sondes at the Canadian stations, from BM sondes at Berlin to GDR sondes and then to ECC sondes at Lindenberg, and from GDR to ECC sondes at Legionowo is indicated by the change in symbol type. The year labels mark the start of the year.

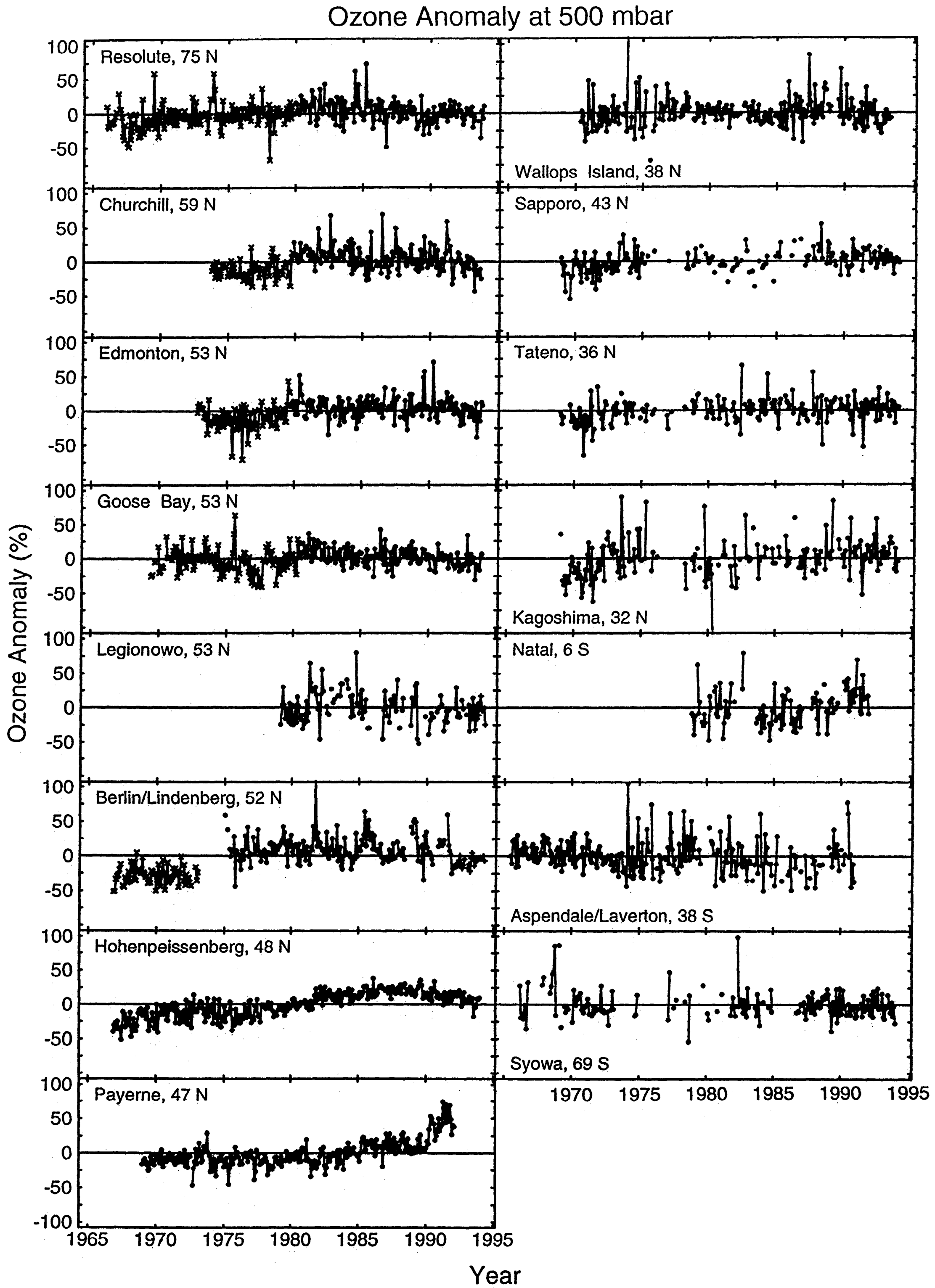


Figure 3b. Monthly mean anomalies at 500 mbar for each ozonesonde station, in percent. See Figure 3a and section 2 for details.

3.2. The Effect of the QBO and Solar Flux Dependence on Trends Derived for Ozone

The QBO signal in the extratropical ozone column lags the QBO in the equatorial zonal wind and is much stronger for winter than for summer [e.g., *Bowman*, 1989]. The optimal QBO lags derived from model 2 for annual and seasonal trends are given in Table 5. The lag tends to decrease with altitude in the stratosphere, from 11 months at 70 mbar to 9 months at 30 mbar at Hohenpeissenberg, for example; the lag near the ozone maximum (~50 mbar for midlatitudes) was used in model 2 for all altitudes. The optimal lag for the annual trend was 13 months for the higher-latitude stations and 10 months for the midlatitude stations, shorter than the lags of 15-16 months reported for the ozone

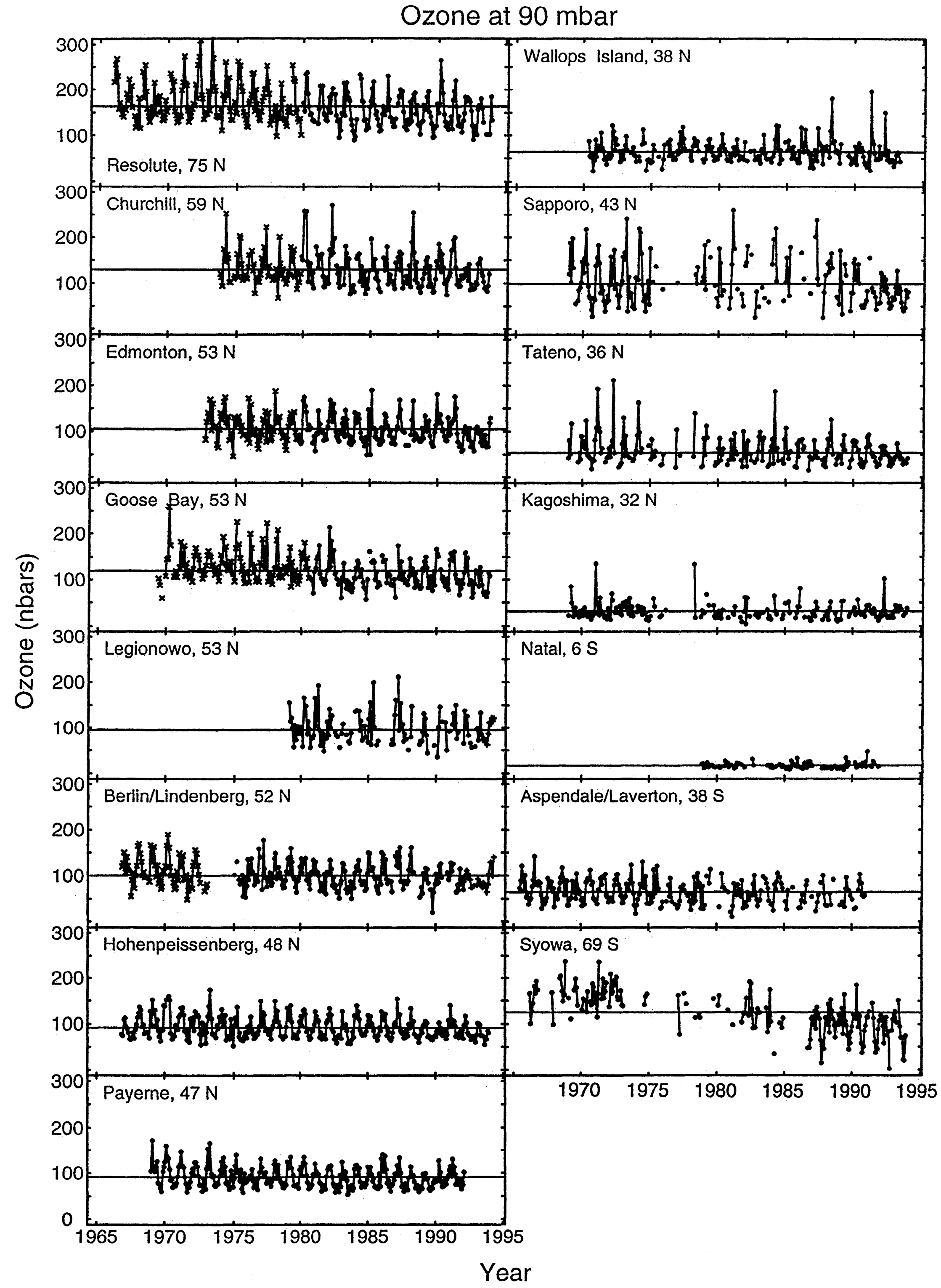


Figure 4a. Monthly mean values of ozone at 90 mbar for each ozonesonde station, given in nanobars. See Figure 3a and section 2 for details.

column at midlatitudes [*Bojkov et al.*, 1990]. *Bowman* [1989] reported that the QBO signal in the midlatitude ozone column was out of phase with that in the tropics, in agreement with the results here.

Inclusion of the dependence of ozone on the QBO and on the solar cycle has very little effect on the magnitude of the trend or the standard error, as illustrated in Figure 7 by results for Hohenpeissenberg. Figure 7 shows trends calculated with models 1 and 2 and the coefficients *b* and *c* in model 2; it includes results for the stations where the differences between models 1 and 2 are largest, Edmonton and Aspendale. Stratospheric ozone at Natal has the strongest dependence on the QBO, consistent with its equatorial location [*Bowman*, 1989; *Zawodny and McCormick*, 1991]. The dependence of ozone on the QBO tends to be significant between

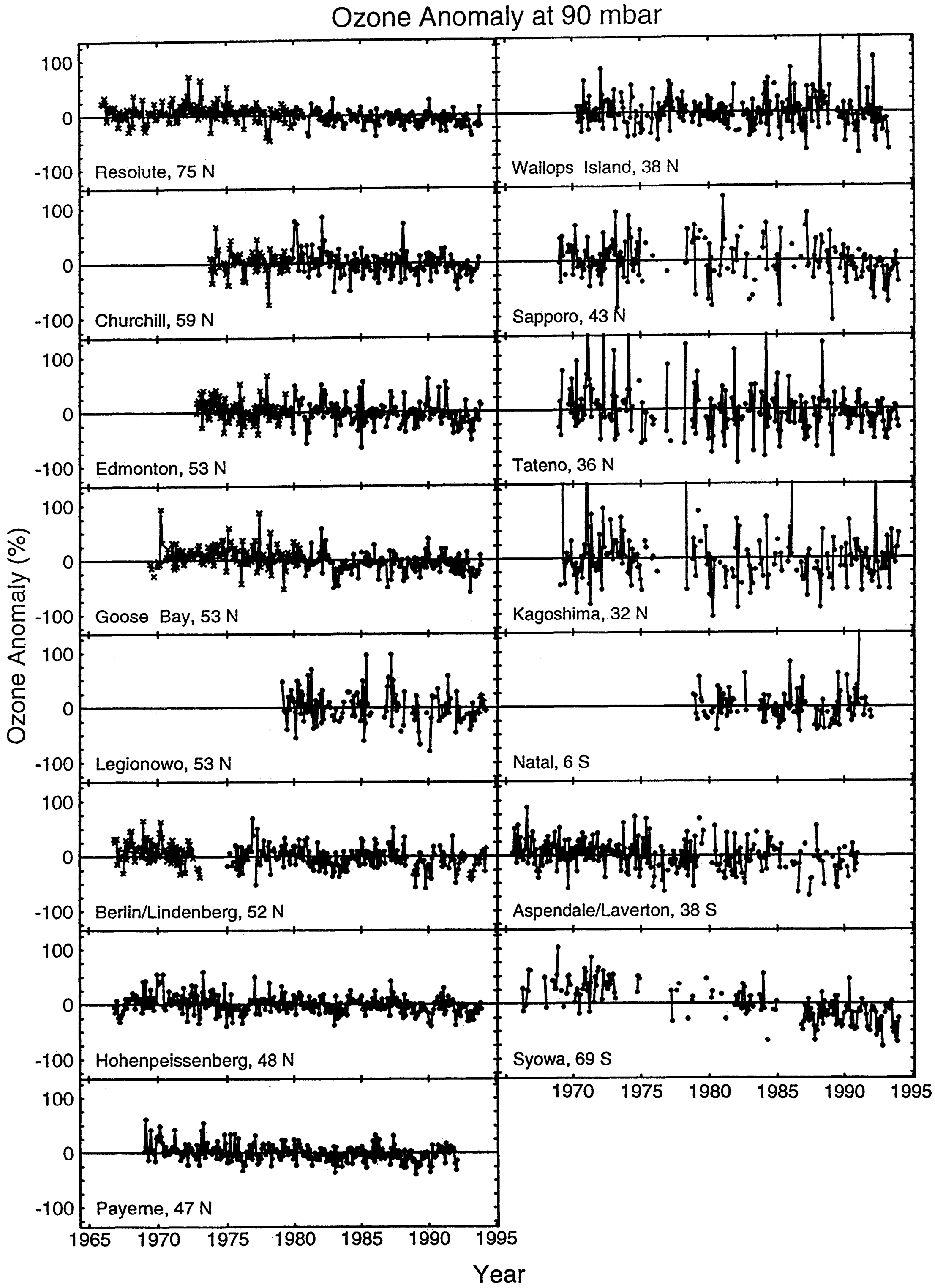


Figure 4b. Monthly mean anomalies at 90 mbar for each ozonesonde station, in percent. See Figure 3a and section 2 for details.

80 mbar and 30 mbar for the midlatitude stations. The dependence of ozone on the 22-year solar cycle is less than that on the QBO for most stations. However, it is dependence on the solar cycle that causes the differences in trends between models 1 and 2 for Edmonton and Aspendale (Figure 7).

The effect of the QBO on ozone is greater in winter than in summer, as shown by the larger and more significant values of b (Figure 8). The optimal lag for the annual trend is usually the same as that for the winter (December-March) trend, since the winter data dominate the QBO signal. The lag for summer (May-August) is usually longer than that for winter [cf. *Bowman*, 1989]. Model 1 was adopted for most of the analyses here, since inclu-

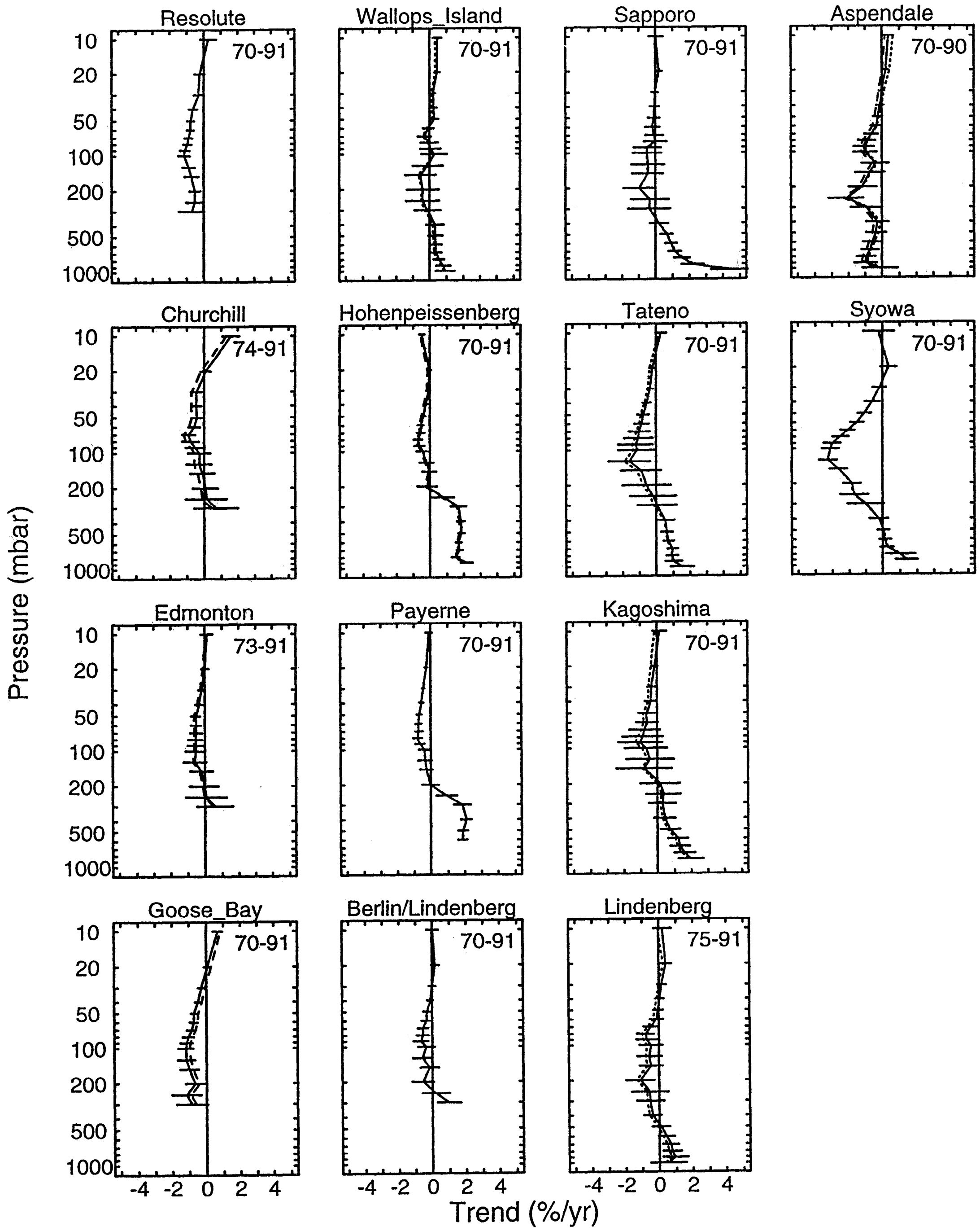


Figure 5. Trend in the vertical distribution of ozone, in percent per year, from the early 1970s to 1991. The years of data included in the analysis are given in the upper right corner of each panel. The solid line shows results obtained by using the data without modification; the horizontal lines show the trend ± 2 standard errors. The long dashed line shows the effect of adjusting the data for several stations to the revised ozone columns using the ratios in Table 3. The short dashed line shows the effect of dividing the data for each sounding by its correction factor prior to analysis (omitted for the Canadian stations and Payerne). Results below 300 mbar are excluded for the Canadian stations because of artifacts introduced by the change in sonde type (see text). Results below 600 mbar are excluded for Payerne because of artifacts introduced by changes in the launch time of the sondes [*Staehelin and Schmid*, 1991]. All data for the months of June, July, and August were omitted for the Japanese stations because of the sparseness of data.

sion of the QBO and solar cycle dependence has little effect on the annual and seasonal trends or their standard errors for most stations.

3.3. Trends in Tropospheric Ozone

Trends in the troposphere are discussed by region, as the magnitude and the vertical distribution of trends vary regionally. Possible causes for the variability in the tropospheric trends are discussed in section 4.

Europe. Hohenpeissenberg and Payerne show increases of about 2% yr^{-1} below 10 km (300 mbar) for 1970-1991 (Figure 5). It is a coincidence that the trends for the two stations are so similar, as the temporal behavior of ozone appears to be different (Figure 3). Ozone values were similar at 500 mbar from 1969 to about

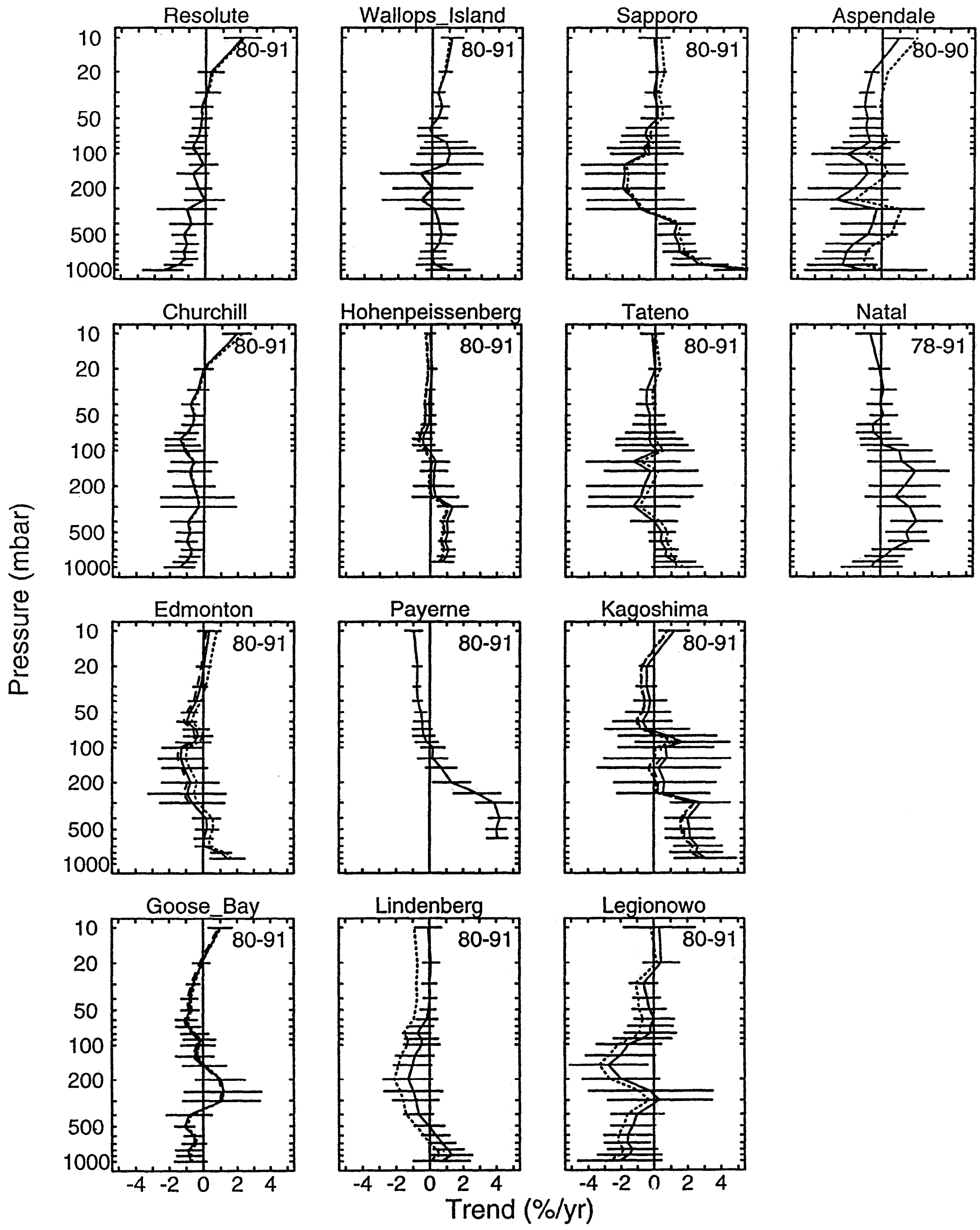


Figure 6. Trend in the vertical distribution of ozone in percent per year, from 1980 to 1991 except where noted in the upper right corner. See Figure 5 for details. Results for Goose Bay are from August 1980 to limit the analysis to ECC sondes.

1979 but were higher at Hohenpeissenberg from 1982 to 1989, and higher at Payerne in 1990 and 1991 (Figure 9). Much of the increase for Hohenpeissenberg occurs between 1977 and 1982, while that for Payerne occurs after 1984, with exceptionally high values in 1990 and 1991 (Figures 3 and 4). There is an increase of ~1% yr^{-1} for 1970-1980 and for 1980-1991 at Hohenpeissenberg (Figures 6 and 10). The increase is significant from the ground to 5.5 km (500 mbar) for the early period, while it is significant up to 10 km (300 mbar) for the later period. There is no increase in ozone at Payerne for 1970-1980 (Figure 6).

The Payerne data are currently being revised, with the use of laboratory calibrations performed since 1984. Preliminary results suggest that the increases shown here after 1983 and the high values in 1990 and 1991 may not be realistic (J. Staehelin, personal communication, 1994), and they are not discussed further.

The temporal behavior of ozone at Hohenpeissenberg since 1978 is similar to that at the nearby (42 km) surface sites of Zugspitze (~3 km) and Wank (~1.8 km) [*Scheel et al.*, 1994]. These sites also show the largest increase for 1978-1982, and the trend for 1978-1991, +1.6% yr^{-1}, is the same as that for Hohenpeissenberg at 700 mbar (3 km). The increase at Zugspitze has leveled off in recent years [*Scheel et al.*, 1993]. Ozone is increasing in all seasons at Hohenpeissenberg, as shown in Figure 11a for 1970-1991 (see also Table 6). Although the percentage increase

Table 5. Optimals Lags for Fit to QBO

Station	Latitude, °N	Year	December-March	May-August
Churchill	59	13	10	16
Edmonton	53	13	13	4
Goose Bay	53	10	10	16
Lindenberg	52	11	11	18
Hohenpeissenberg	48	10	10	2
Payerne	47	10	10	12
Sapporo	43	0(10)	0(10)	NA
Wallops Is.	38	10	10	16
Tateno	36	7	-1	NA
Kagoshima	32	10	10	NA
	Latitude, °S	Year	July-October	November-March
Natal	6	-1	-2	-1
Aspendale	38	12	12	-13

The table shows the optimal lag (in months) that gives the best fit to model 2 for 50 mbar (30 mbar for Natal); a range of lags from -15 to 25 was tested. For Sapporo, the fit with lag 0 is only slightly more significant than the fit with lag 10. NA indicates not applicable, because of so many missing data.

is largest in autumn and winter, the absolute increase is about the same year round, ~1 ppb yr^{-1}. At the Zugspitze station the increase is higher in summer than in winter, ~1.3 and 0.7 ppb yr^{-1}, respectively, for median values in 1978-1990 [*Scheel et al.*, 1994].

The trends at Hohenpeissenberg since 1980 are sensitive to the period chosen, because ozone has been decreasing in the last few years (Figure 3 and Table 6). The trend for 1980-1993 is smaller than that for 1980-1991, and is significant only in the lower troposphere (Figure 12). There is no trend since 1982, except in the surface data (Tables 6, 7 and 8). There are inconsistencies between the surface data and the sonde data for 900 mbar (close to the surface, 975 m), in terms of both ozone concentrations and

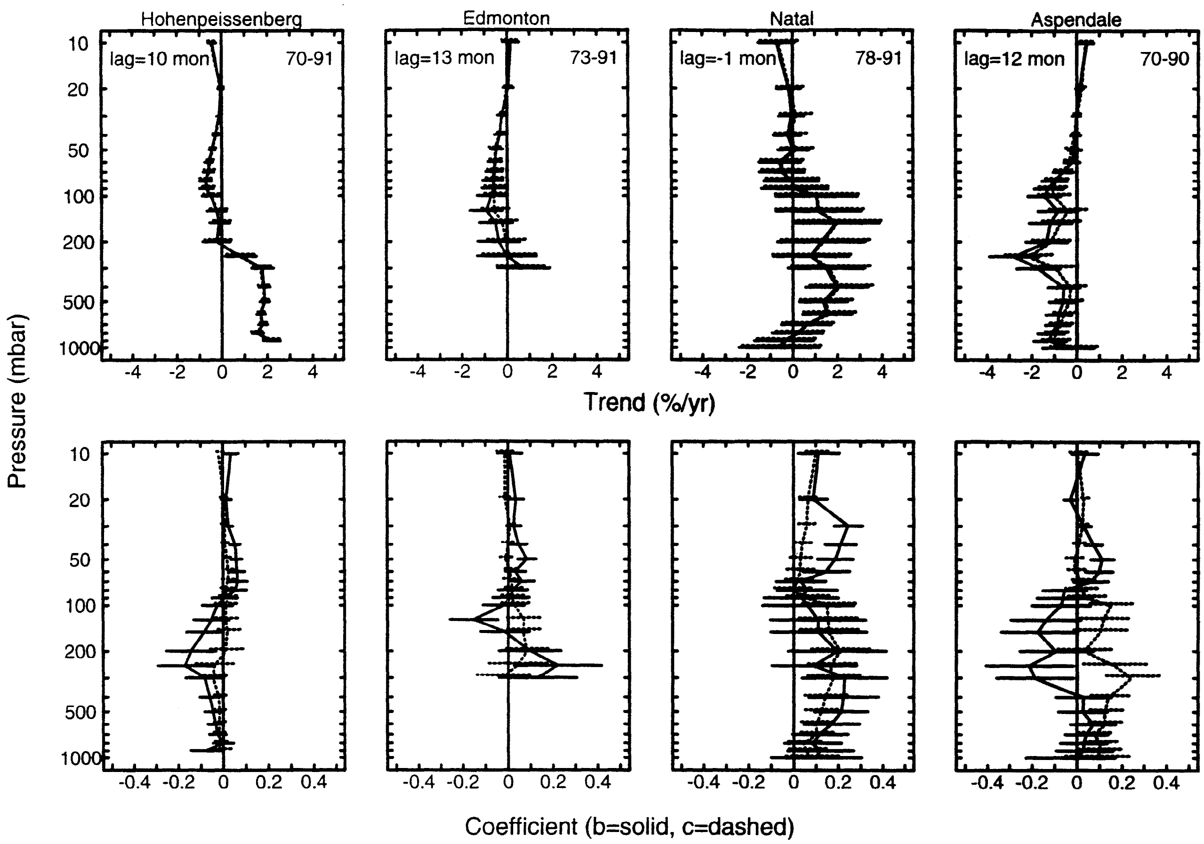


Figure 7. Trend in the vertical distribution of ozone in percent per year, calculated with model 2 (solid line), which allows for the dependence of ozone on the QBO and solar flux (see section 2.2); the dashed line shows the trends for model 1. The top panels show the trends, and the lower panels give the coefficients *b* and *c* in equation (2). Two standard errors are given for the trends and for *b* and *c*. The lag used for the QBO dependence is given in the upper left. For the stations not shown, the results for models 1 and 2 were almost indistinguishable.

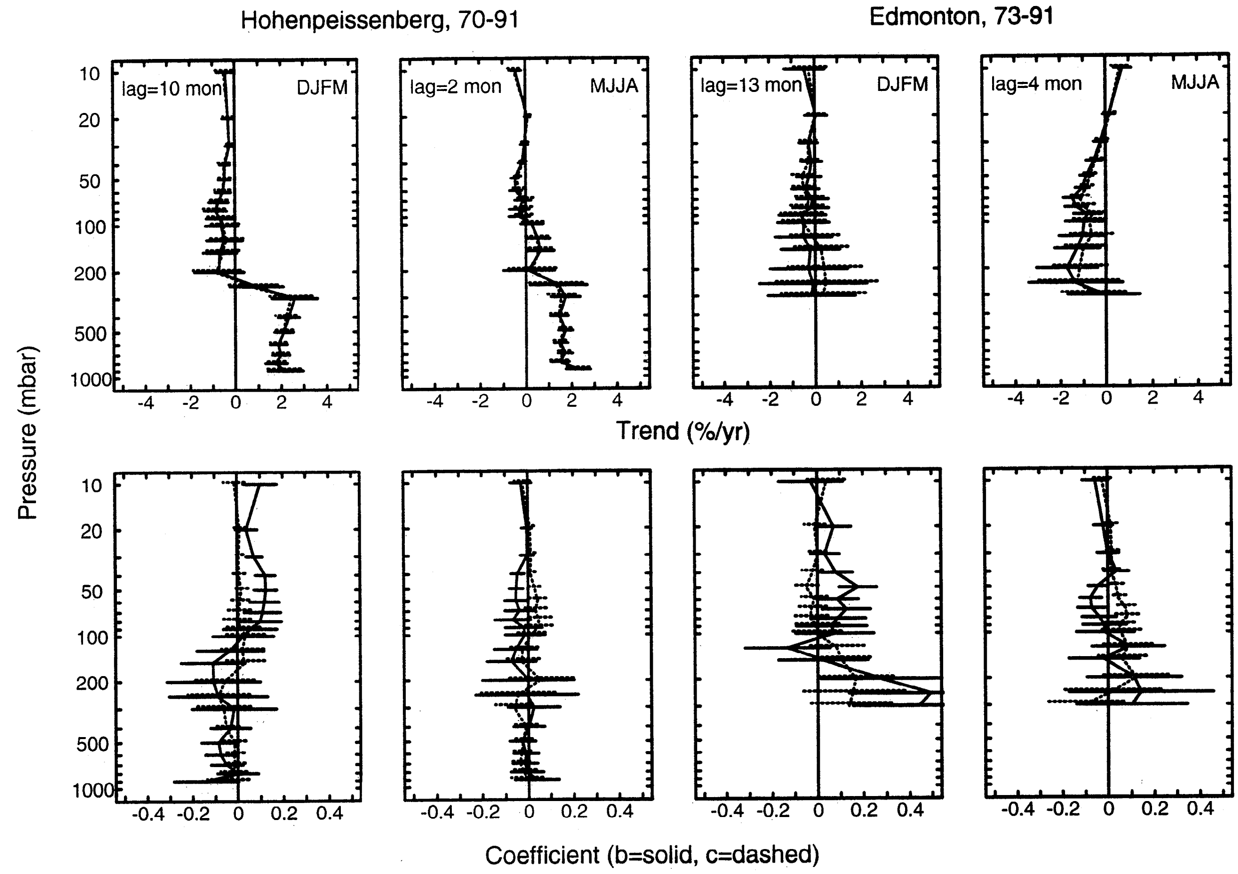


Figure 8. Trend in the vertical distribution of ozone in percent per year, calculated with model 2 (solid line) and model 1 (dashed line), for December-March and May-August (see Figure 7). Different lags were used for the QBO dependence for each season, as discussed in the text.

trends (Figure 13). Although the sondes are launched near the time of the minimum in the diurnal variation of surface ozone (around 0700 [*Bojkov*, 1988]), the values recorded are closer to the surface means than to the surface minima. This suggests nighttime depletion of ozone in the surface layer, by deposition or by titration with NO_x. Trends in the surface data are larger than those in the sonde data for 900 mbar after 1980 (Table 7) but are smaller than those for the sonde data for 1976-1992 (Table 8); these differences are statistically significant. The largest increase in the surface data is in the minimum values.

The increase in ozone at Hohenpeissenberg is largest for 900 mbar for 1970-1991. The sonde data may not represent the true change in ozone because of the likely decrease in ambient SO_2; there is a filter for SO_2 on the surface instrument [*Low et al.*, 1991]. Values of ozone at 900 mbar range from a few parts per billion to about 30 ppb in the 1970s in winter; the winter trend is 0.7 ppb yr^{-1}. Winter concentrations of SO_2 in the early 1980s were 5-20 ppb at nearby Garmisch (740 m) and 1-8 ppb at Wank (1780 m), and summer values were 1-5 ppb; concentrations of

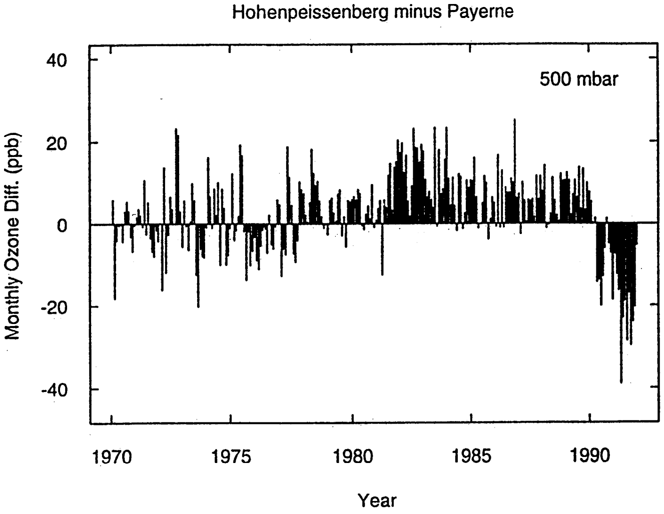


Figure 9. The difference in monthly means values for Hohenpeissenberg and Payerne, in parts per billion, at 500 mbar.

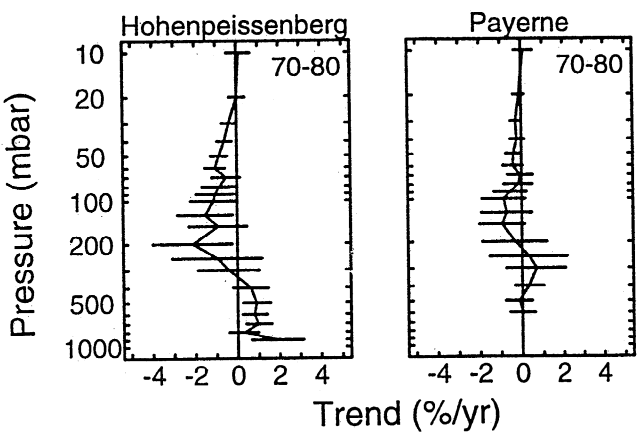


Figure 10. Trend in the vertical distribution of ozone in percent per year, from 1970 to 1980 for Hohenpeissenberg and Payerne.

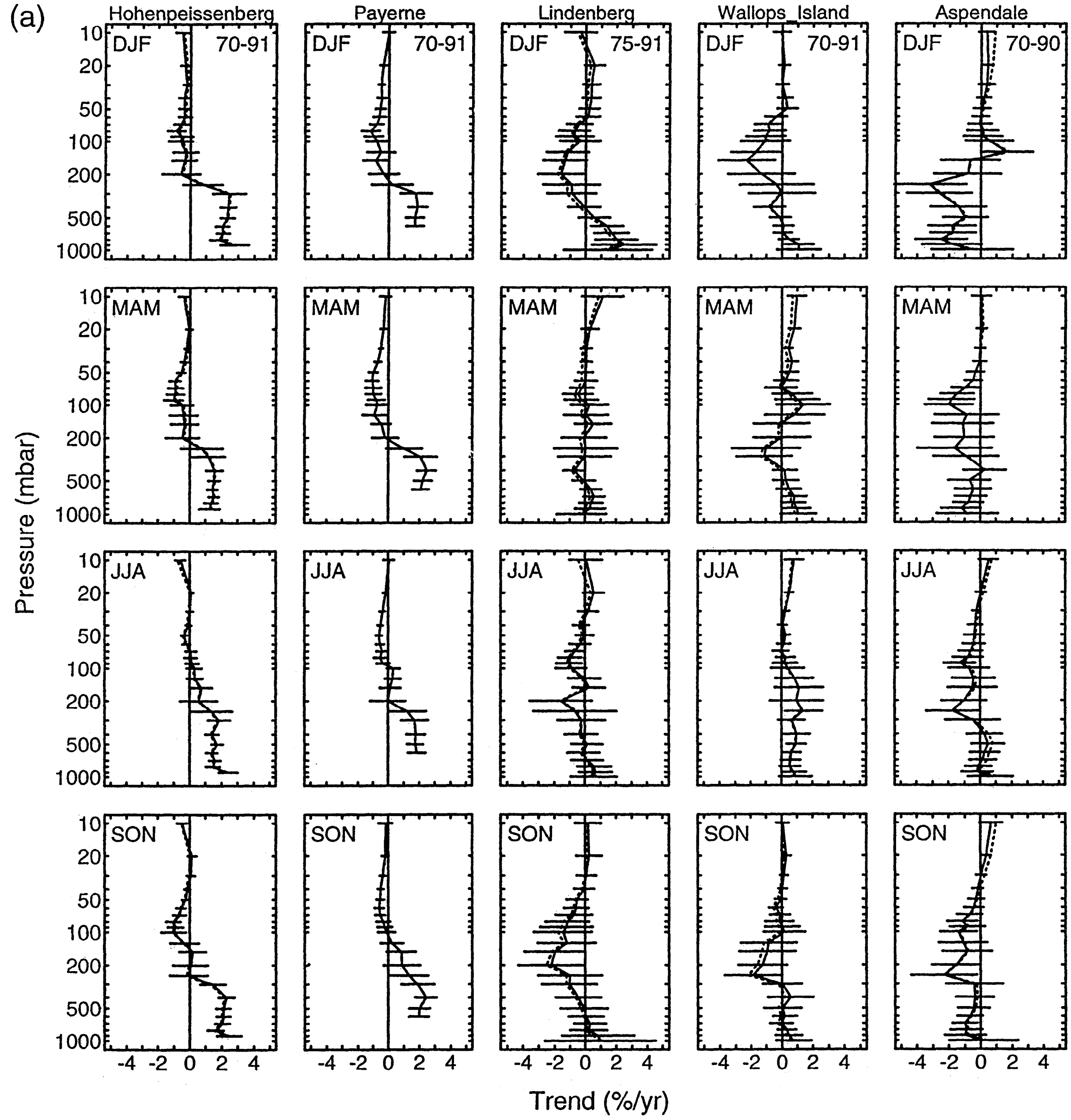


Figure 11a. Trend in the vertical distribution of ozone by season for the early 1970s to 1991, except where noted. The dashed line shows the effect of dividing the data for each sounding by its correction factor prior to analysis. See Figure 5 for further details.

NO_x were somewhat higher [*Reiter et al.*, 1987]. If values of SO_2 at Hohenpeissenberg (975 m) were similar to those at Garmisch or Wank, the trend results for ozone in winter are not correct. Furthermore, concentrations of NO_2 were likely high enough to titrate some of the ozone in winter, so that near-surface trends are not regionally representative. Exposure to high concentrations of pollution could have affected the sonde measurements throughout the troposphere. *Attmanspacher and Dutsch* [1970] reported that BM sondes underestimated ozone the most on days with pronounced inversions (i.e., highest pollution levels) during the intercomparison at Hohenpeissenberg in January 1970. Trends derived for the lower troposphere in winter may well be overestimates.

Furrer et al. [1992] reported an increase in tropospheric ozone of about 2% yr^{-1} for Berlin/Lindenberg. They did not address the 30% jump in ozone values (Figure 3) caused by the change in sonde type when the station was moved to Lindenberg, and this shift appears to be the major contributor to their trend. For the Lindenberg data alone, there is an increase in ozone of ~2% yr^{-1} only in winter below 3 km (700 mbar), and no trend in other seasons (Figure 11a). Contamination by SO_2 may affect the quality of the sonde data, since concentrations of ozone reported for 1000 mbar are often low, <15 ppb in November to March. Emissions of SO_2 in GDR increased by 36% from 1975 to 1987 and then started to decline [*OECD*, 1993], so the effect of SO_2 would be to mimic a decrease in ozone, the opposite of what is observed; the cause of the winter increase in ozone is unclear. Data are lacking to evaluate the extent of NO_x titration in winter. There is a large trend in the correction factors for Lindenberg after 1980 (Table 2), and there are concerns about the quality of the column data used to scale the sondes (section 2.1). The GDR sondes used at Linden-

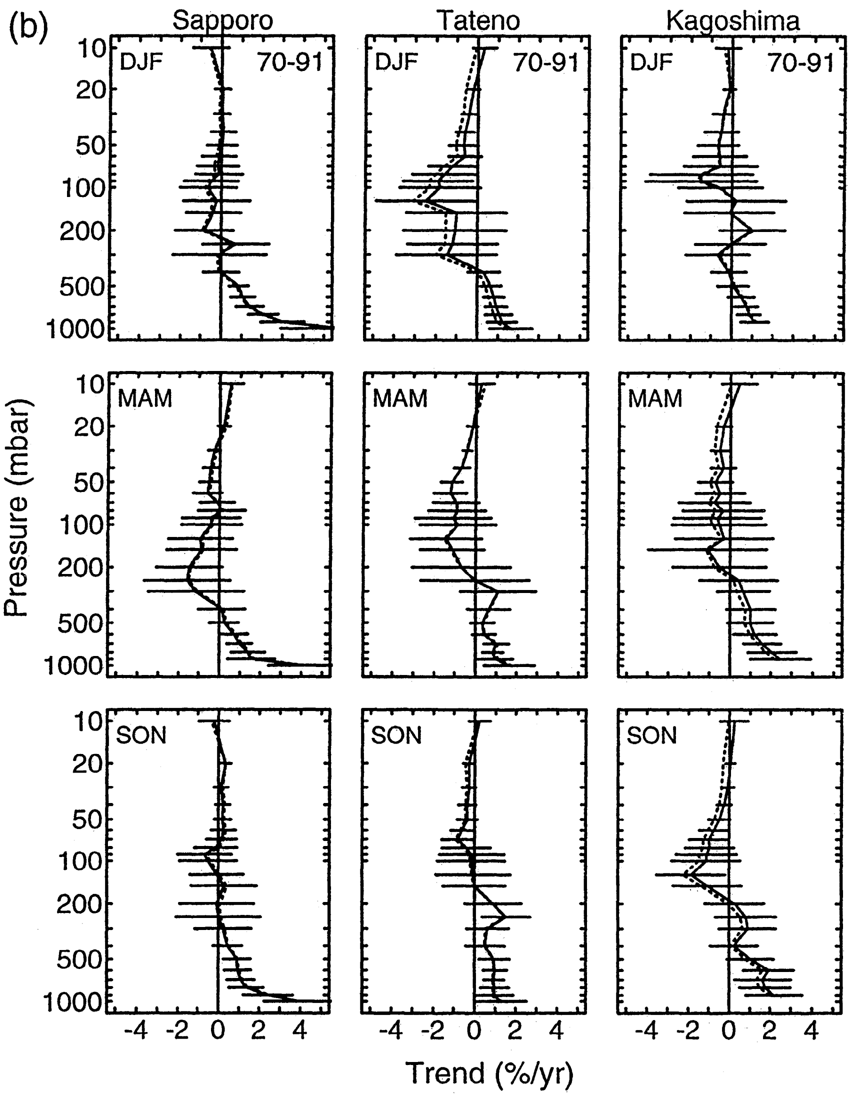


Figure 11b. See Figure 11a. Data are unavailable for the Japanese stations in summer.

berg did not perform reliably at the 1978 intercomparison and have not been tested in subsequent intercomparisons. Data taken with GDR sondes at Legionowo have unusually high correction factors (Table 1), and concentrations are highly variable (Figure 3). ECC sondes have been used at Lindenberg since March 1992, with a downturn in values reported for the troposphere (Figure 3), another indication that the GDR sondes overestimated tropospheric ozone.

North America. Three of the Canadian stations show decreases in tropospheric ozone for 1980-1991, while Edmonton shows no trend except for 900 and 800 mbar where there is an increase (Figure 6); these results agree with the analysis of *Oltmans* [1993]. Data for 1992 and 1993 support the decrease in tropospheric ozone (Figure 12). Seasonal trends are variable and are often not significant (Table 6). The increase in ozone in the boundary layer (Edmonton is located at 670 m, ~935 mbar) is caused by the change in the time of measurement from early morning to late afternoon for the period 1985-1989; ozone values for sondes released at 1600 hours are 20% larger than those from sondes released at 0400 hours. There is a strong diurnal variation in surface ozone near Edmonton [*Angle and Sandhu*, 1986, 1989].

The question of long-term increases in ozone is addressed in Figure 14, which shows profiles for the first 5 years of data and for 1987-1991 for the Canadian stations; the data were first adjusted to the revised ozone columns (see Table 3). The ratio of the later to the earlier 5-year average is less than 1.2 everywhere, except for 900 and 800 mbar at Edmonton, and often less than 1.15. Given that the first 5 years of data were obtained with BM sondes and the later years with ECC sondes, the latter giving about 15-20% more tropospheric ozone (see section 2), these results show no evidence for a long-term increase in tropospheric ozone. The same conclusion holds when the data are analyzed by season, with the exception of data from Resolute in summer, when the ratio is 1.25-1.3 for 700-900 mbar and <1.2 above.

The comparison in Figure 14 contradicts the results of *Wang et al.* [1993], who show increases of ~1% yr^{-1} for all the Canadian stations. In their study, the change in sonde type was accounted by using ratios derived from concurrent ECC data from Garmisch and BM data from Hohenpeissenberg (R. D. Bojkov, personal communication, 1993); *Bojkov* [1988] states that these data show that ECC sondes give 4-12% more tropospheric ozone than BM sondes. About 200 soundings were launched on the same day at

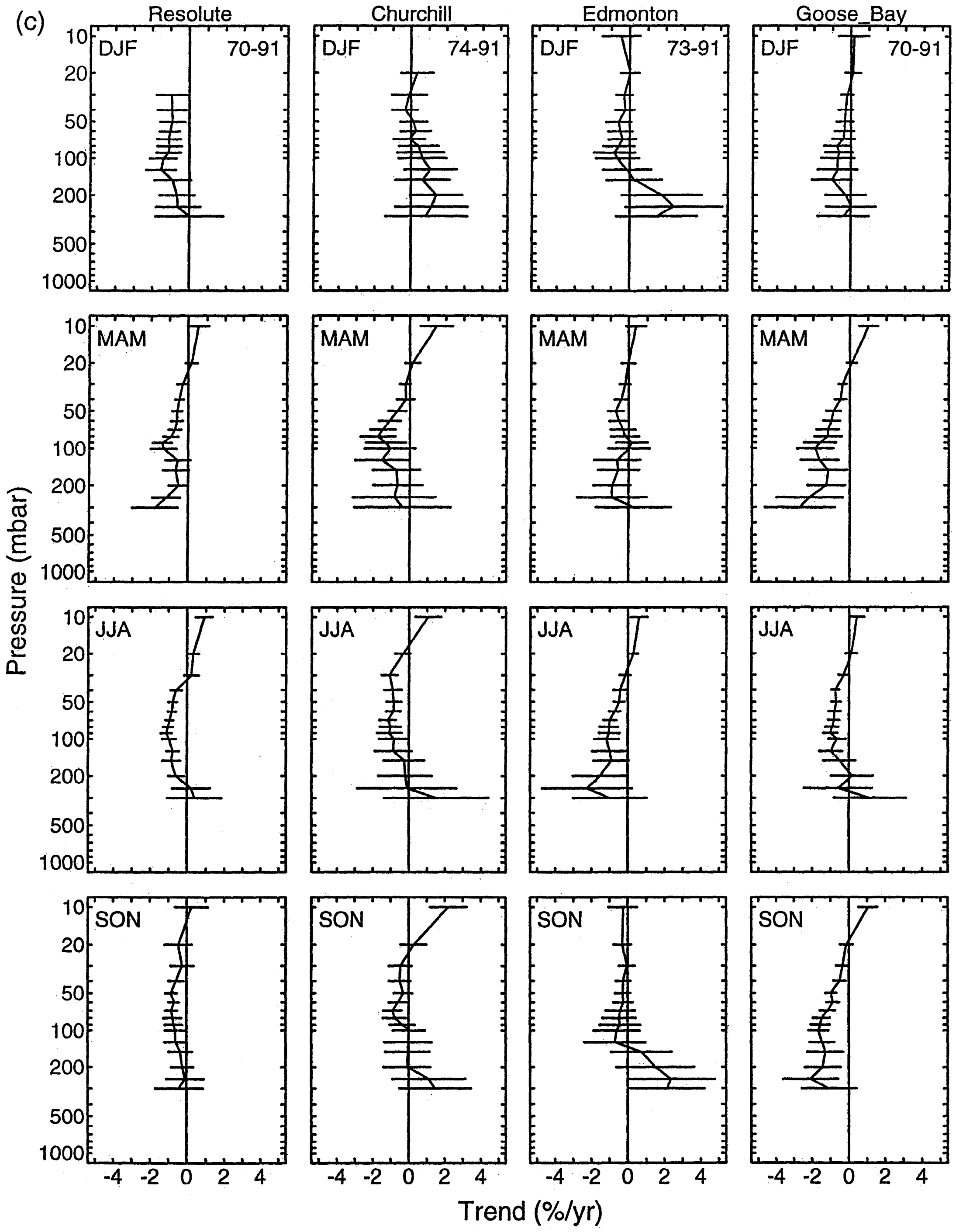


Figure 11c. See Figure 11a.

the two locations between 1978 and 1985. Figure 15 shows the difference in ozone measured by the two sonde types at 500 mbar for 113 pairs of soundings; these soundings had been scaled to identical ozone columns and had corrections factors of 0.8-1.2 and 0.9-1.2 for ECC and BM data, respectively. The figure shows that the ECC sondes gave more ozone than BM sondes in 1978-1979, about the same amount in 1980-1981, and less ozone than the BM sondes in 1982-1985. Similar results were found for other pressure levels in the troposphere. Given the drift of the ECC values relative to the BM values, the data from Hohenpeissenberg and Garmisch cannot be used to relate measurements of tropospheric ozone made by the two types of sondes. BM sondes from Hohenpeissenberg gave less tropospheric ozone than ECC sondes in the two intercomparisons conducted in 1978 and 1984, as discussed in section 2, suggesting a drift in the Garmisch data.

London and Liu [1992] and *London* [1994] report long-term increases for the Canadian stations but mention no adjustment for the instrument change, thus introducing an artificial increase into their results.

Wallops Island, on the east coast of the United States, is the only North American station to show a long-term annual increase in ozone in the midtroposphere, and it is small, <0.5% yr^{-1}, and significant only below 4 km (600 mbar, Figure 5). The increase is slightly less than 1% yr^{-1} below 10 km (300 mbar) in summer for

Table 6. Seasonal Trends in Ozone at 500 mbar yr^{-1}

Station	Years	DJF	MAM	JJA	SON
Resolute	1980-1991	-1.6 ± 1.4	(-1.3 ± 1.5)	(-1.0 ± 1.2)	(-1.2 ± 1.5)
	1980-1993	-1.1 ± 1.1	-1.6 ± 1.2	-1.0 ± 1.0	-1.2 ± 1.2
Churchill	1980-1991	-1.2 ± 1.2	(0.7 ± 1.4)	(-0.6 ± 1.7)	-2.8 ± 1.3
	1980-1993	(-0.7 ± 0.9)	(-0.4 ± 1.2)	-1.4 ± 1.4	-2.6 ± 1.1
Edmonton	1980-1991	-0.7 ± 0.8	(0.4 ± 1.4)	(0.6 ± 1.6)	(0.5 ± 1.2)
	1980-1993	-0.6 ± 0.6	(-0.6 ± 1.1)	(-0.1 ± 1.3)	(0.1 ± 0.9)
Goose Bay*	1980-1991	(-1.8 ± 1.4)	(-0.8 ± 1.0)	(-0.9 ± 1.2)	-1.8 ± 1.1
	1980-1993	-1.7 ± 1.1	-0.9 ± 0.7	-1.5 ± 0.9	(-0.9 ± 1.0)
Wallops Island	1970-1991	(-0.1 ± 0.7)	(0.3 ± 0.8)	0.9 ± 0.6	(0.0 ± 1.2)
	1970-1992	(0.0 ± 0.6)	(0.1 ± 0.1)	(.57 ± .63)	(-0.1 ± 1.1)
	1980-1991	(0.3 ± 1.2)	(-0.3 ± 2.0)	1.3 ± 1.2	(0.7 ± 2.3)
	1980-1992	(0.6 ± 1.1)	(-0.7 ± 1.7)	(0.2 ± 1.3)	(0.1 ± 2.0)
Lindenberg	1975-1991	(0.6 ± 1.0)	(-0.1 ± 0.7)	(0.1 ± 1.0)	(-0.1 ± 1.5)
	1980-1991	(0.6 ± 1.5)	(0.0 ± 1.1)	(0.7 ± 1.9)	(-1.5 ± 2.8)
Hohenpeissenberg	1970-1991	2.3 ± 0.5	1.5 ± 0.4	1.7 ± 0.4	2.1 ± 0.4
	1970-1980	(1.4 ± 1.6)	(0.5 ± 1.2)	(0.6 ± 1.1)	(1.2 ± 1.6)
	1980-1991	1.0 ± 0.7	1.0 ± 0.7	1.3 ± 0.7	(0.7 ± 0.7)
	1980-1993	(0.4 ± 0.6)	0.6 ± 0.6	(0.3 ± 0.7)	(0.0 ± 0.6)
Payerne	1970-1991	1.7 ± 0.6	2.2 ± 0.6	1.7 ± 0.5	2.0 ± 0.7
	1980-1991	3.7 ± 1.3	3.5 ± 1.1	4.3 ± 0.9	4.5 ± 1.4
Sapporo	1970-1991	0.9 ± 0.5	(0.2 ± 0.7)	ND	(0.9 ± 0.7)
	1980-1991	1.4 ± 1.0	(0.9 ± 2.0)	ND	(1.1 ± 1.9)
	1980-1993	1.0 ± 0.7	(0.6 ± 1.4)	ND	(0.5 ± 1.4)
Tateno	1970-1991	0.7 ± 0.5	(0.3 ± 0.6)	ND	0.9 ± 0.8
	1980-1991	(0.0 ± 1.0)	(1.0 ± 1.4)	ND	(0.1 ± 1.6)
	1980-1993	(0.5 ± 0.8)	(0.4 ± 1.0)	ND	(-0.2 ± 1.2)
Kagoshima	1970-1991	(0.1 ± 0.8)	(1.0 ± 1.1)	ND	(1.0 ± 1.1)
	1980-1991	(1.2 ± 1.6)	2.9 ± 2.6	ND	(1.9 ± 2.6)
	1980-1993	1.1 ± 1.1	2.2 ± 1.9	ND	(0.7 ± 1.9)
	1981-1993	(0.9 ± 1.2)	(0.6 ± 1.5)	ND	(0.5 ± 2.0)
Aspendale	1970-1990	(-1.0 ± 1.3)	(-0.7 ± 1.3)	(0.5 ± 1.0)	(-0.4 ± 1.0)
	1980-1990	(-2.8 ± 4.4)	(-1.7 ± 3.9)	(1.8 ± 3.6)	(-1.7 ± 2.2)

The trend was calculated by using the residuals for the months indicated. The table shows the trends and twice the standard error, in percent per year. Trends given in parentheses are not statistically significant. ND indicates no data.

*Values for Goose Bay start in September 1980.

1970-1991, and it is smaller and not significant if data for 1992 are included (Table 6). There is no significant trend in other seasons except below 3 km in spring and below 1 km in winter (Figure 11a). There is no significant annual trend for 1980-1991 (Figure 6), and the summer trend is significant for 500 mbar only; there is no significant trend in summer for 1980-1992 (Table 6). Interference from SO_2 may be a problem for Wallops Island in the boundary layer in winter.

Ozone profiles were measured over Boulder by using BM sondes in 1963-1966 [*Dutsch et al.*, 1970] and ECC sondes in 1985-1989 [*Oltmans et al.*, 1989]. Figure 16 shows a comparison of monthly mean values for each period, along with the BM data multiplied by 1.15 at 500 and 700 mbar to allow for the different tropospheric response of the two techniques, discussed above. There appears to have been an increase in ozone only at 700 and 800 mbar; these pressure levels are in the boundary layer at Boulder, which is at an altitude of 1650 m (~830 mbar). The increase is likely caused by an increase in local pollution from Boulder and nearby Denver. There is no evidence for a long-term increase in tropospheric ozone at 500 or 300 mbar (5.5-9 km).

Japan. The Japanese stations show increases of <1% yr^{-1} for 5.5 km (500 mbar) for 1970-1991, with larger increases at lower altitudes (Figures 5 and 6); the increase is present in all seasons with data (Figure 11b). Similar results were found by *Akimoto et*

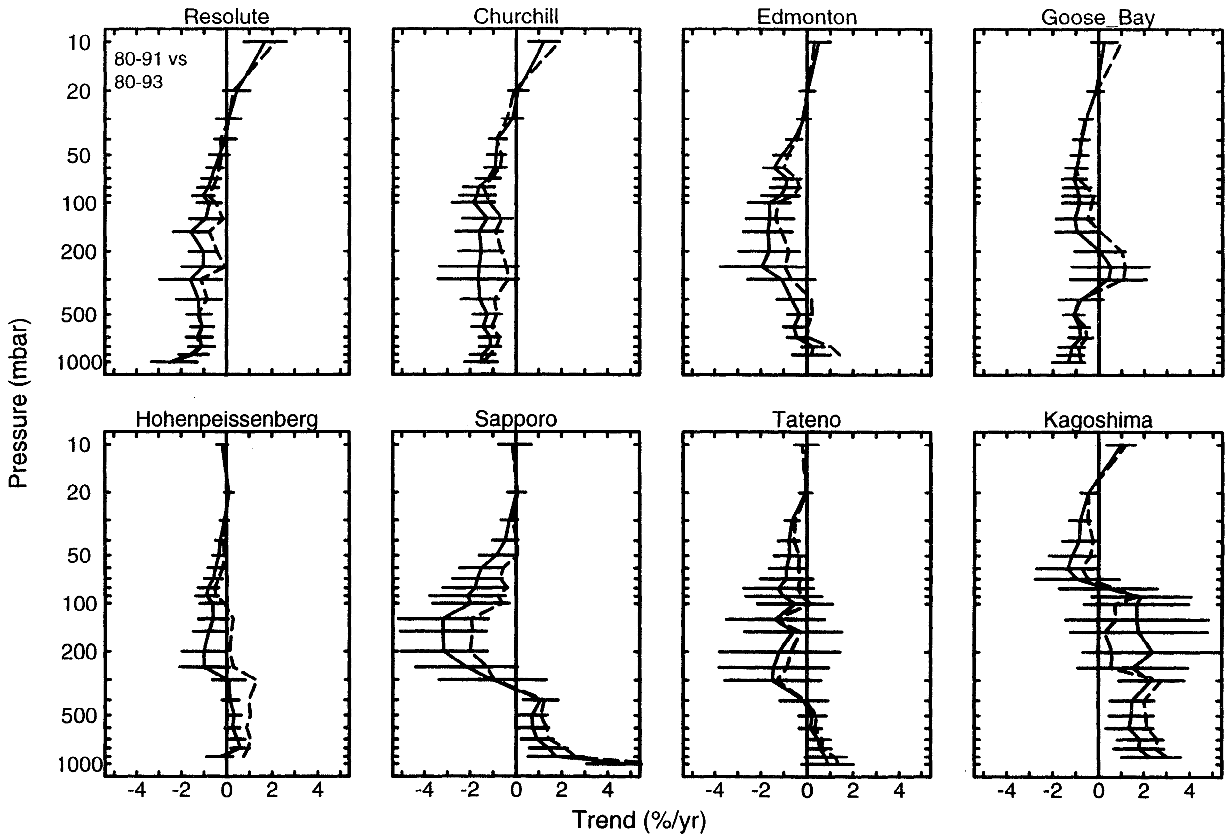


Figure 12. Trend in the vertical distribution of ozone in percent per year, for 1980-1993 (solid) compared to 1980-1991 (dashed). Two standard errors are shown for the trends for 1980-1993.

al. [1993]. For the period 1980-1991, the increase is about the same for Tateno, larger for Sapporo, which has a sparse data record in the mid-1980s (Figure 3), and much larger for Kagoshima. Results for Kagoshima are sensitive to low values for 1980; trends starting in 1981 are smaller (Table 6). Trends for 1980-1993 tend to be smaller than those for 1980-1991 (Figure 12 and Table 6). Trends below 2 km are unusually large at Sapporo, where surface concentrations are often below 5 ppb in winter; values for other months are also unusually low. The low values and large trend may be a result of high levels of SO_2 interfering with the ozone measurement, or they could reflect titration by high levels of NO_x. None of the Japanese stations shows a significant trend for 300 mbar for 1970-1991 or 1970-1993, although Kagoshima gives a significant trend for 1980-91.

Natal, a tropical station with 13 years of sparse data, shows an increase in ozone in the middle and upper troposphere, but the trend is significant only for 400-600 mbar (Figures 6 and 7). There is a small decrease in ozone, <1% yr^{-1}, for 600-800 mbar at Aspendale/Laverton, the only midlatitude station with a long record in the southern hemisphere (Figure 5). The trend is largest in the austral summer (Figure 11a) and may be related to the decrease in stratospheric ozone at high southern latitudes. There is no trend for 400-700 mbar at Syowa, Antarctica, but there is an increase of 1-1.5% yr^{-1} for 800 and 900 mbar. By contrast, data

Table 7. Trends in Ozone at Hohenpeissenberg

Years	Surface	900 mbar	700 mbar	500 mbar	300 mbar
1980-1991	1.4 ± 0.5	0.7 ± 0.6	1.1 ± 0.4	1.0 ± 0.3	1.3 ± 0.9
1980-1992	1.2 ± 0.5	(0.3 ± 0.6)	0.8 ± 0.4	0.7 ± 0.3	(0.6 ± 0.8)
1982-1991	1.5 ± 0.7	(-0.4 ± 0.8)	(0.2 ± 0.4)	(0.2 ± 0.4)	(0.6 ± 1.2)
1982-1992	1.2 ± 0.6	(-0.7 ± 0.7)	(0.1 ± 0.4)	(0.0 ± 0.4)	(-0.2 ± 1.0)

The table gives the trend and twice the standard error, in percent per year. Trends given in parentheses are not statistically significant at the 95% confidence level.

Table 8. Trend in Ozone at Hohenpeissenberg, 1976-92

	Annual	DJF	MAM	JJA	SON
900 mbar	1.7 ± 0.5	3.1 ± 1.6	1.4 ± 0.9	1.8 ± 0.9	1.6 ± 1.6
Surface mean	0.9 ± 0.3	1.5 ± 1.0	(0.8 ± 0.8)	0.9 ± 0.8	(0.5 ± 0.8)
minima	1.9 ± 0.5	4.2 ± 2.0	1.7 ± 0.8	1.3 ± 0.9	1.8 ± 1.2
maxima	(0.0 ± 0.4)	(-0.7 ± 1.9)	(-0.2 ± 0.8)	(0.7 ± 0.8)	(-0.3 ± 1.0)

The table gives the trend and twice the standard error, in percent per year. Trends given in parentheses are not statistically significant at the 95% confidence level.

from the South Pole indicate a decrease in surface ozone of 0.7% yr^{-1} since 1975 [*Oltmans and Levy*, 1994].

3.4. Stratospheric Trends

The vertical distribution of the trend in stratospheric ozone is shown in Figures 5, 6, and 11. Most of the sonde stations in the northern hemisphere show a maximum decrease of -0.7 to -1.2% yr^{-1} near 90 mbar (17 km) from the early 1970s until 1991. The decrease in ozone extends from about 30 mbar to near the tropopause, encompassing the ozone maximum. The change in sonde type in Canada could have introduced a small (at most ~ -0.2% yr^{-1}) artifact in the results for 1970-1991, as BM sondes give at most 5% more ozone than ECC sondes (section 2.1). The results in Figure 6 are in qualitative agreement with trends derived from SAGE I and SAGE II for February 1979 to April 1991, which show a decrease in ozone for 17-24 km (90-30 mbar), but the SAGE data give a larger trend, about -1.6% yr^{-1}, for ~90 mbar at midlatitudes [*McCormick et al.*, 1992].

The SAGE data, above 17 km, give column trends about 20% smaller than TOMS [*McCormick et al.*, 1992]. The sonde data indicate that loss of ozone from 250 mbar to 90 mbar (10-17 km) could account for the difference between TOMS and SAGE; Table 9 gives the fraction of column loss of ozone in the strato-

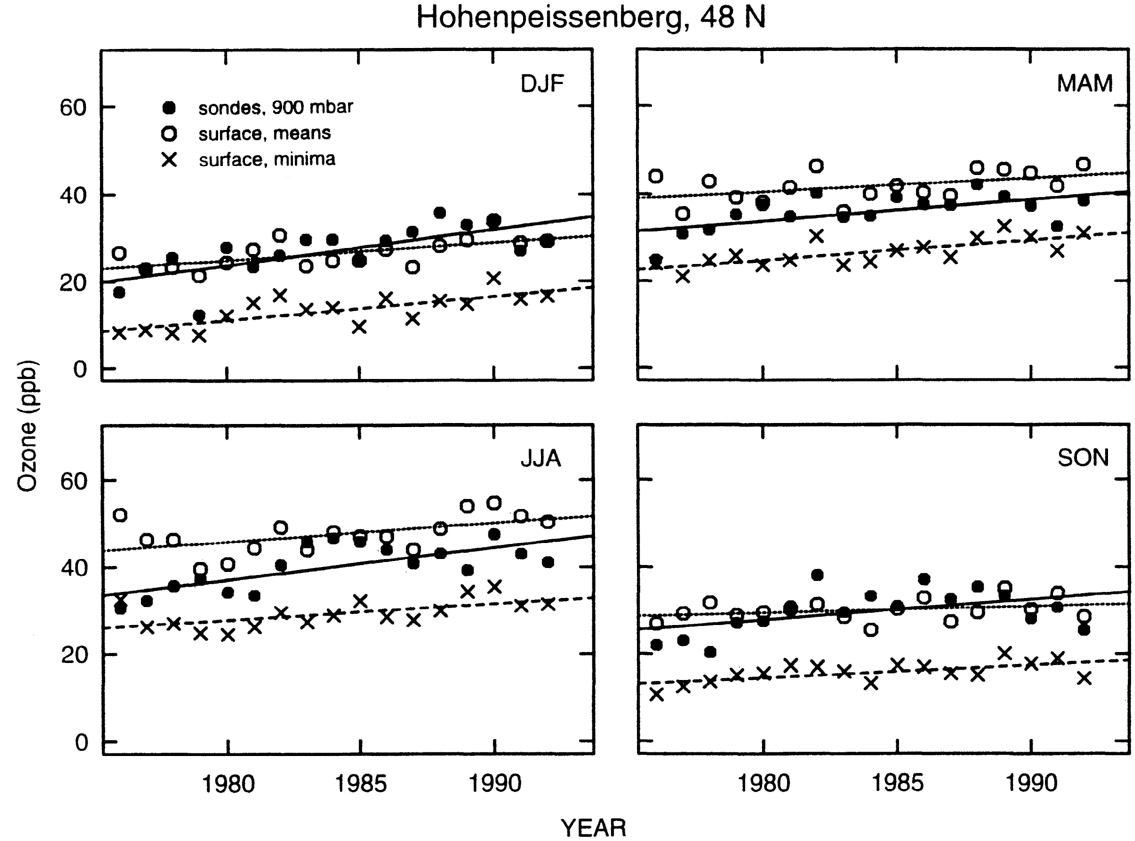


Figure 13. Trend in ozone at the surface and at 900 mbar for Hohenpeissenberg, shown as seasonal means. The open circles show the average of the daily means of the surface data, and the crosses show the average of the daily minima of the surface data. The solid circles show the sonde data at 900 mbar, near the surface. The lines show the trend for 1976 to 1992, for each set of data.

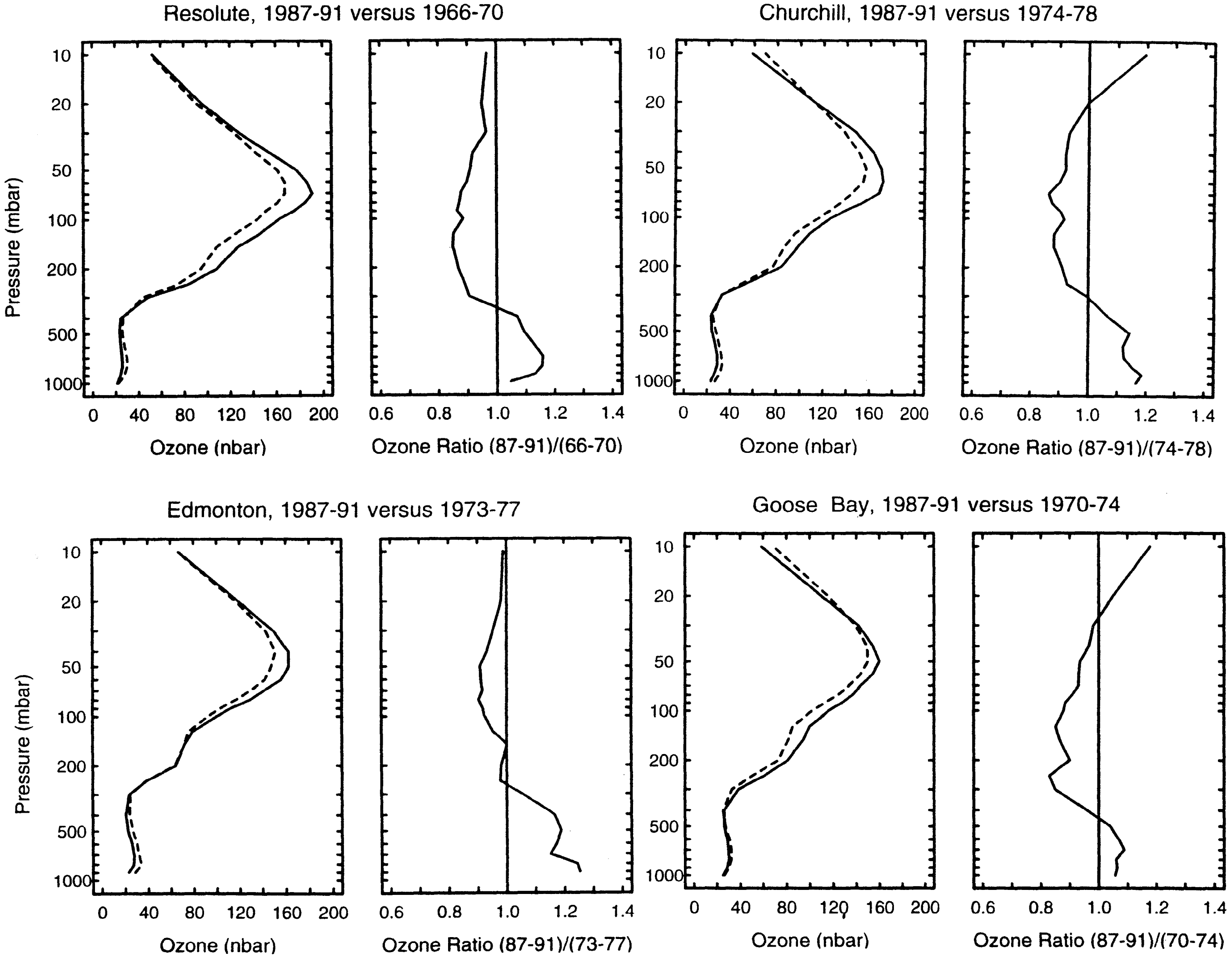


Figure 14. Comparison of mean values for first 5 years of data and for 1987-1991 for the Canadian stations. The sonde data were first adjusted to the revised column data by using the ratios given in Table 3. The left panels show concentrations for each time period, and the right panels show the ratio of the concentrations for the two periods.

sphere that occurs below 17 km, for the ozone trends in Figure 11. Loss of ozone below 17 km (90 mbar) accounts for 15-50% of the column loss of stratospheric ozone for the stations with more frequent measurements.

Decreases in ozone in the lower stratosphere are larger for 1980-1993 than for 1980-1991 (Figure 12). This is a consequence of unusually low values of ozone in the lower stratosphere in 1992-1993 following the eruption of Pinatubo in June 1991 [e.g., *Kerr et al.*, 1993; *Hofmann et al.*, 1994]. Low values are evident at 90 mbar in 1992-1993 for most of the sonde stations for which data are available (see Figure 4).

Trends in stratospheric ozone derived from the sonde data are qualitatively similar to trends derived from ozone column measurements, both ground-based and TOMS [*Stolarski et al.*, 1992], and show similar regional variability. The sonde data are scaled to the ground-based column data, so consistency is expected; however, the sonde trends appear consistent with the column data even when the scaling is removed prior to analysis (short dashed lines in Figures 5, 6, and 11), with the exceptions of Lindenberg and Aspendale. Trends for December to March and for May to August, the seasons adopted by *Stolarski et al.* [1992], are shown in Figure 17 for 1979 to 1991.

The decrease in ozone derived from the sonde data is largest in spring and smallest in summer for Hohenpeissenberg and Payerne for 1970-1991 (Figure 11). For the 4-month seasons, the trend is larger in winter than summer for all four European sonde stations (Figure 17), a pattern seen also in TOMS.

Ozone is decreasing in all seasons for the Canadian stations for 1970-1991, with the exception of Churchill in winter (Figure 11c). There are problems with the winter column data for Churchill [*Bojkov et al.*, 1988] that are not removed by the procedure used here to adjust the sondes to the revised column data. For 1979-1991 the decrease is larger in summer than in winter for all four stations (Figure 17). The TOMS data also show larger trends in summer for Goose Bay and Churchill, and about the same trend in summer as for winter for Edmonton.

The sonde data for Wallops Island give a significant decrease only in winter (Figure 11a) for 1970-1991, in agreement with the column data. The winter trend for 1979-1991 is smaller than that for any other sonde station, a pattern not seen in TOMS. There is no trend in summer in the sonde data or in the ground-based column data, but TOMS shows a small decrease. Comparison of data from Boulder for 1963-1966 with data for 1985-1989 suggests maximum ozone loss of about -0.8 to -1.0% yr^{-1} at 100 mbar

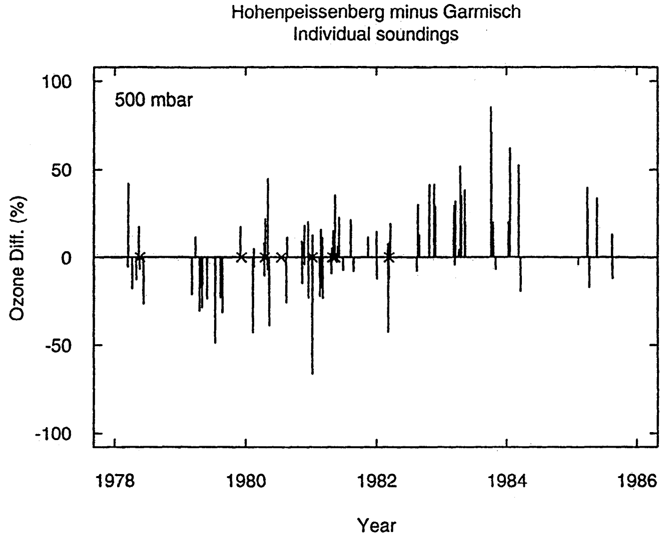


Figure 15. The difference between ozone measured by BM sondes from Hohenpeissenberg and by ECC sondes from Garmisch, in percent. Results are shown for 500 mbar, for 113 pairs of soundings launched on the same day that were scaled to the same value for the ozone column; BM soundings had a corrections factors of 0.9-1.2, ECC values of 0.8-1.2. The crosses indicate pairs of soundings that gave identical values for ozone.

in winter and -0.3 to -0.6% yr^{-1} at 60 mbar in summer (J. A. Logan et al., manuscript in preparation).

The short record from Natal, from late 1978 until 1991, shows no significant trend in the lower stratosphere; the record is sparse and does not provide an adequate test of the tropical trends obtained from SAGE I and II for 1979-1991 [*McCormick et al.*, 1992]. Aspendale shows a decrease in stratospheric ozone in all seasons except austral summer (Figure 11a and Figure 17). The column data show a similar decrease in summer and in winter [*Stolarski et al.*, 1992]. This discrepancy may reflect either the low sampling frequency of the sondes or problems with data quality.

The most dramatic decreases in stratospheric ozone are seen for Syowa, on the Antarctic coast [*Chubachi and Kajiwara*, 1986; *Iwasaka and Kondoh*, 1987]. Figure 18 compares ozone from the two periods with most frequent measurements, before 1974 and after 1986. Ozone is 60-75% lower at the ozone maximum in the later period during September to December, and there are decreases of over 30% in most other months. The decrease occurs over a greater vertical extent (20-300 mbar) in austral spring than in other seasons, when it occurs primarily below 50 mbar. The altitude of maximum ozone loss decreases from ~17 km (70 mbar) in September to ~12.5 km (150 mbar) in February. The ozone values reported for 90 mbar before 1974 show little overlap with those obtained after 1986 in the months of greatest ozone loss (Figure 19). *Oltmans et al.* [1994] compared vertical profiles over the South Pole for similar time periods. They also found significant ozone losses in austral spring and summer but found no loss in winter, in contrast to the losses year round over Syowa. They noted a September minimum in the stratosphere prior to the onset of the ozone hole. There is a hint of such a minimum over Syowa (Figure 20).

4. Discussion

4.1. Stratospheric Ozone

Trends derived from ozonesondes support the major conclusions of the recent WMO assessment [*WMO*, 1992] that stratospheric ozone is decreasing in seasons other than winter at middle and high latitudes and that most of the decrease is occurring in the lower stratosphere. The WMO report relied primarily on ozone column measurements and SAGE data and considered results from only one sonde station, Payerne. The sonde data analyzed here

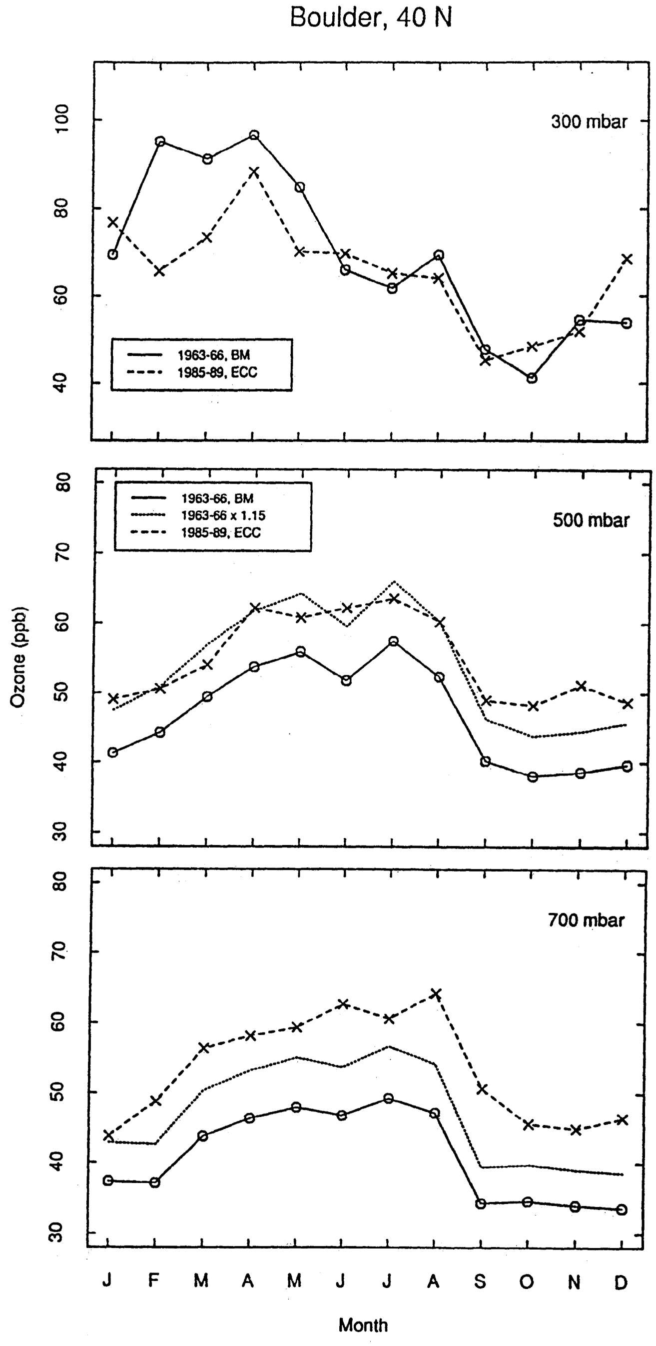


Figure 16. Seasonal distribution of tropospheric ozone over Boulder in 1963-1966 (dashed lines, BM sondes) and 1985-1989 (solid line, ECC sondes). The dotted line (lower two panels) shows the data from 1963-1966 multiplied by 1.15.

Table 9. Fraction of Ozone Loss in Lower Stratosphere

Station	Years	DJF	MAM	JJA	SON
Resolute	1970-1991	0.37	0.46	0.38	0.24
Churchill	1974-1991	0.00	0.39	0.16	0.00
Edmonton	1973-1991	0.13	0.43	0.48	0.30
Goose Bay	1970-1991	0.45	0.48	0.22	0.40
Lindenberg	1970-1991	0.69	0.37	0.17	0.23
	1975-1991	0.72	0.00	0.48	0.56
Hohenpeissenberg	1970-1991	0.26	0.21	0.00	0.21
Payerne	1970-1985	0.24	0.21	0.00	0.00
Sapporo	1970-1991	0.80	0.56	ND	0.00
Tateno	1970-1991	0.34	0.25	ND	0.00
Kagoshima	1970-1991	0.00	0.18	ND	0.18
Aspendale	1970-1991	0.00	0.38	0.34	0.52

The table gives the fraction of ozone loss below 17 km (90 mbar). This was calculated by integrating the column loss of ozone from 90 mbar down to the lowest level of ozone loss above 300 mbar, and from 25 mbar down to the same level, and taking the ratio. The change in ozone above 25 mbar was generally small and highly uncertain (see Figure 11). ND indicates no data.

show that ozone losses occur down almost to the tropopause, particularly in winter and spring, and that loss below 17 km (~90 mbar) can account for the smaller column ozone losses derived from SAGE than from TOMS. A recent analysis of Canadian data for January to April 1993, when record low values of ozone occurred, also found ozone loss down to 10 km (~250 mbar) [*Kerr et al.*, 1993]; similar results were found for Boulder for the winter/spring of 1992-1993 [*Hofmann et al.*, 1994].

The increase in tropospheric ozone over Hohenpeissenberg makes a large contribution to the trend in column ozone, about +1.5%/decade for 1970-1991, of opposite sign to column trend caused by stratospheric loss. The average column loss of ozone for midlatitudes over this period is -1.5 to -2.1%/decade [*Stolarski et al.*, 1992; *Reinsel et al.*, 1994]. The spatial extent of the increase over Europe is unclear; there is no evidence for a similar increase over Lindenberg, 500 km to the north. For other sonde stations, the contribution of the tropospheric trend to the column change is less than 1.0%/decade and is often insignificant (Table 10).

Stratospheric ozone loss is larger in winter and spring than summer for the sonde stations in Europe (47°-53°N) and the United States (~40°N). A similar pattern is observed in the ground-based and column data for 30°-50°N [*Stolarski et al.*, 1992; *Niu et al.*, 1992; *Reinsel et al.*, 1994]. For the Canadian stations, located between 53°N and 75°N, the summer loss is as large as (or larger than) the winter loss, similar to results for the column data.

Two-dimensional models of stratospheric chemistry that include a representation of heterogeneous processes on sulfate aerosols predict about the same loss of ozone in summer as in winter during the 1980s, as chlorine levels increase. The models predict about the correct column loss of ozone in summer and loss of about -0.5% yr^{-1} in the lower stratosphere at midlatitudes; they underpredict winter loss [*WMO*, 1992]. Three-dimensional models will be needed to investigate the mechanisms responsible for the spatial patterns in ozone loss seen in the sonde and in the column data.

4.2. Tropospheric Ozone

Tropospheric ozone is produced by photochemical oxidation of CO, CH_4, and hydrocarbons in the presence of NO_x [e.g., *Fishman et al.*, 1979b]. Nitrogen oxides are the limiting species for ozone production in most of the troposphere because of their low concentrations, typically less than 100 ppt [e.g., *Chameides et al.*, 1992; *Carroll and Thompson*, 1993]; this is the case even in rural areas of the eastern United States, where values are of the order of 1 ppb in summer [*Trainer et al.*, 1987; *Sillman et al.*, 1990; *Seinfeld et al.*, 1991]. Trends in tropospheric ozone are discussed here in the context of trends in emissions of NO_x from fossil fuel combustion in the boundary layer ("surface sources") and from aircraft. Aircraft emissions, while a small fraction of combustion-related sources of NO_x, have significant potential to affect tropospheric ozone because of the nonlinear dependence of

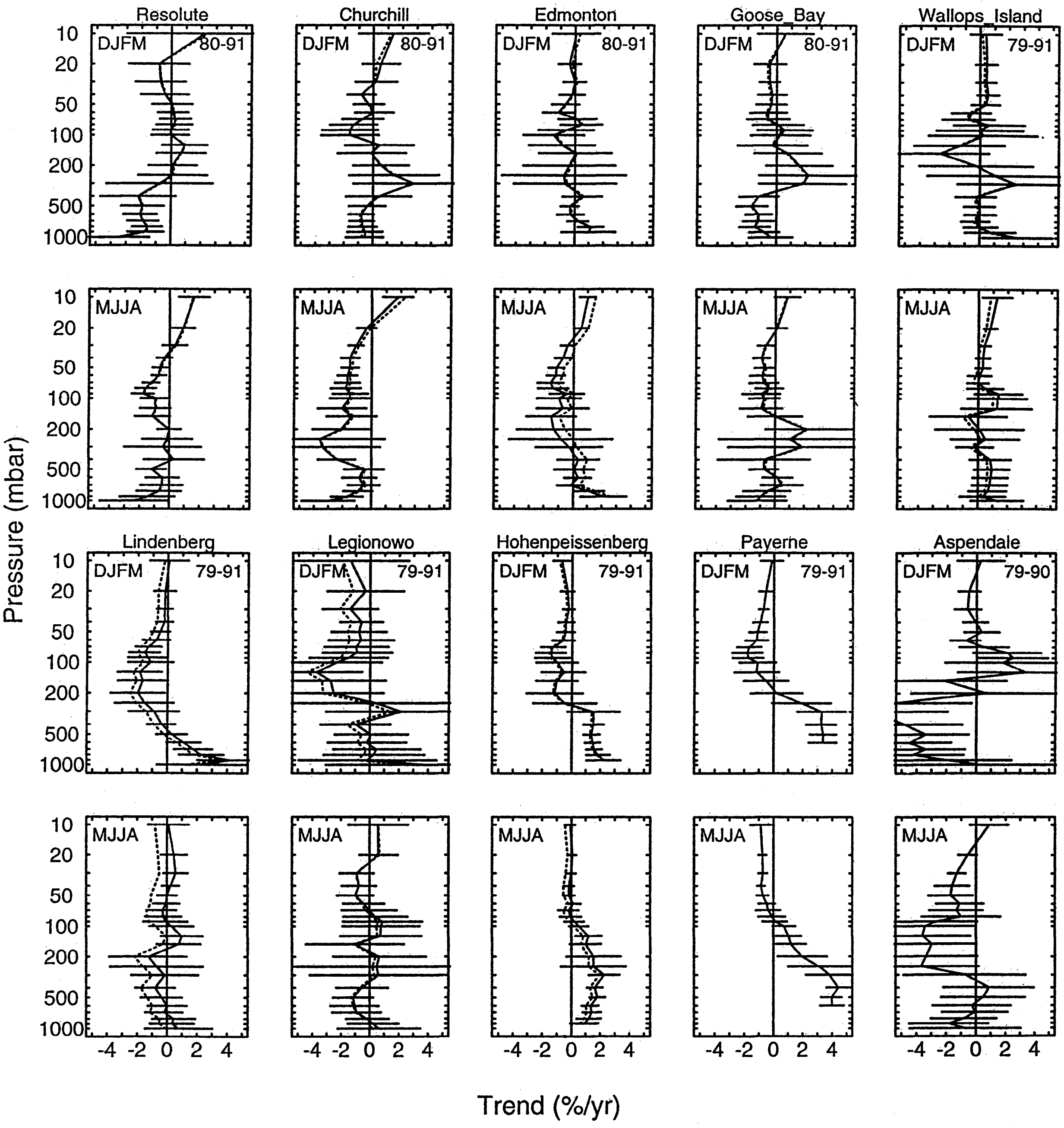
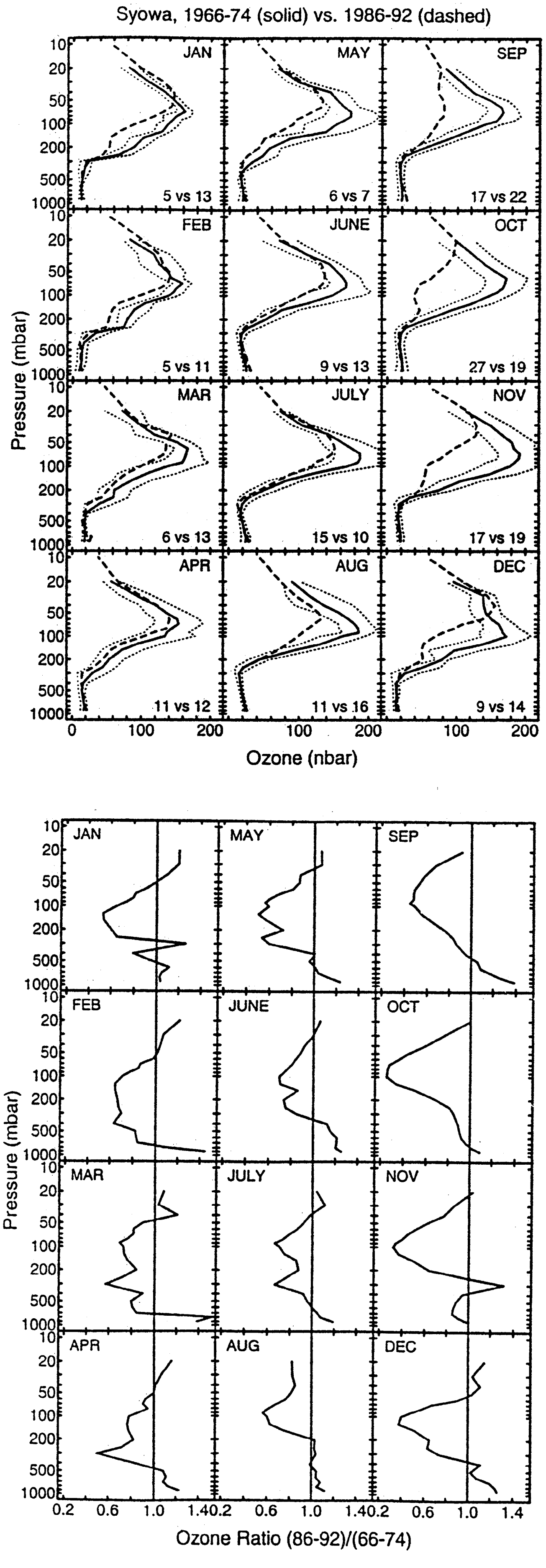


Figure 17. Trend in the vertical distribution of ozone for the 4-month seasons used in the analysis of TOMS for 1979-1991, December-March and May-August. Results for the Canadian stations are for 1980-1991 and use only ECC sondes. Results for Aspendale are for 1979-1990. The dashed line shows the effect of dividing the data for each sounding by its correction factor prior to analysis.

ozone production on NO_x concentrations. The ozone production efficiency, the amount of ozone produced for each molecule of NO_x emitted, is largest (40-80) for the low concentrations found in remote air and decreases to less than 10 for NO_x concentrations greater than 1 ppb [*Liu et al.*, 1987].

Surface emissions of NO_x increased by 20% in the 1970s in the United States and by 27% in Western Europe and have remained approximately constant since (Figure 21). Urban measurement of NO_2 provides the only test of trends in emissions; measurements from ~200 sites in the United States show a downward trend of ~0.5% yr^{-1} in the 1980s [EPA, 1989, 1992b]. Sparser data from Europe show that there have not been major changes in emissions in the 1980s, but do not provide a stringent test of the trends [*Eggleston et al.*, 1992; *OECD*, 1993]. Data from Japan suggest an increase in emissions rather than the decrease shown in Figure 21 [*Japan Environmental Agency*, 1989; *OECD*, 1993; *Gotoh*, 1993]. There was a substantial increase in emissions only in Asia (excluding Japan), about 50% in 10 years [*Kato and Akimoto*, 1992]. Global emissions of NO_x from fossil fuel use were 73 Tg (as NO_2) in 1985 [*Benkovitz*, 1993]. The increase in Asia represents a hemispheric increase of 6% from 1975 to 1985, assuming that 90% of global emissions are in the northern hemisphere.

Emissions of NO_x from aircraft were 1.5 Tg in 1990 [*Wuebbles et al.*, 1993], a factor of 3 greater than an estimate for 1975 [*Bauer*, 1982]; the reliability of the earlier estimate is unknown.



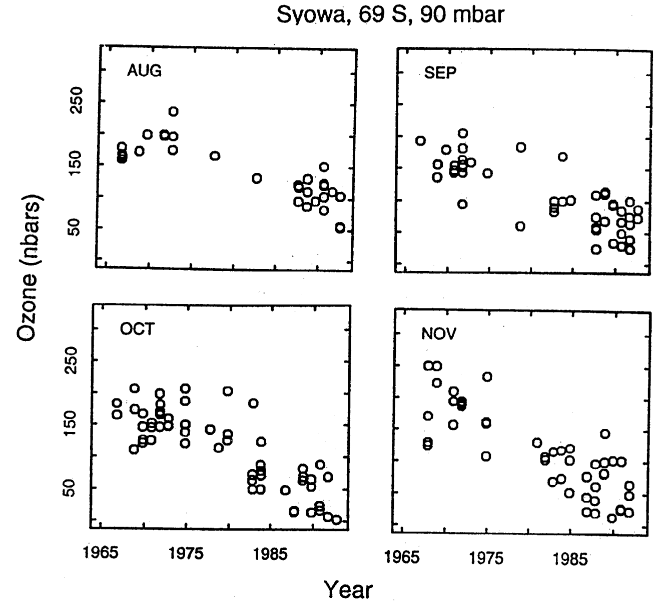


Figure 19. Concentration of ozone (nanobars) at 90 mbar for each ozone sounding at Syowa in the months of August to October.

Global consumption of jet fuel doubled over the past 20 years (Figure 22). Jet fuel use increased 17% in the United States and 55% in western Europe in the 1970s and increased by about 35% in both regions in the 1980s [*International Energy Agency* (hereinafter IEA), 1987, 1991, 1993]. The density of aircraft emissions in 1990 is slightly higher over the eastern United States than over western Europe and smaller over eastern Asia and Japan; emissions peak at about 10-11 km, or ~250 mbar [*Wuebbles et al.*, 1993]. A significant fraction of aircraft emissions, over 40%, are estimated to be injected into the stratosphere in winter and spring, but in summer almost all the emissions are in the troposphere [*WMO*, 1992].

Estimates of the effects of increases in NO_x emissions on tropospheric ozone have been made only with two-dimensional (2-D) models. These studies suggest that aircraft emissions of NO_x are more effective than surface sources in increasing tropospheric ozone [*Johnson et al.*, 1992]. The models give a range of ozone increases of 3-12% between 350 and 200 mbar for aircraft emissions of 1.5-2.0 Tg NO_2 [*Beck et al.*, 1992; *Johnson et al.*, 1992; *WMO*, 1992]; these results imply an increase in ozone of 0.1-0.5% yr^{-1} if scaled to account for an increase of 1.0 Tg NO_2 over 15 years. Two-dimensional models are not, however, suitable tools for quantifying the effect of NO_x emissions on ozone [e.g., *Thompson*, 1992]. They do not include convective processes that mix surface sources of NO_x out of the boundary layer, and they cannot treat the inhomogeneities in the NO_x distribution and the resulting nonlinearities in the chemistry of ozone.

Preliminary results from a three-dimensional (3-D) model for North America suggest that about 6% of the NO_x from fossil fuel

Figure 18. Comparison of the first 9 years (solid line) and last 7 years (dashed line) of data for Syowa; the dotted lines show the mean ± 1 standard deviation, for the early period. The top panels show concentrations for each time period for each month; the number of soundings is given for 1966-1974 (left) and 1986-1992 (right) for each month. The lower panels show the ratio of the concentrations for the two periods.

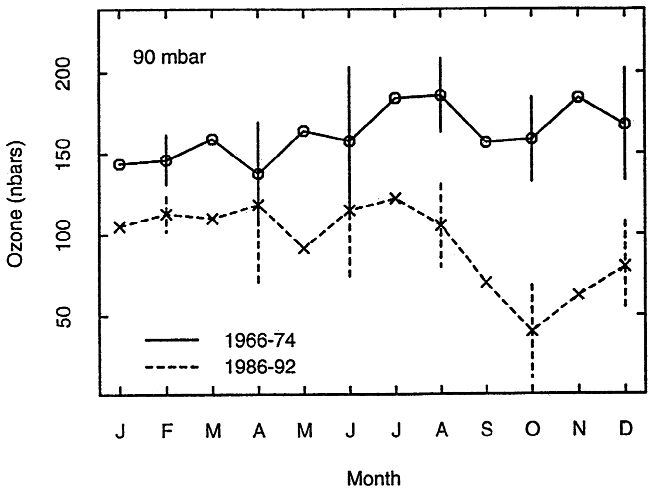


Figure 20. Seasonal cycle of ozone (in nanobars) at Syowa at 90 mbar, for 1966-1974 (solid line) and 1986-1992 (dashed line). The vertical bars show one standard deviation.

combustion is exported from the boundary layer in summer as NO_x [*Jacob et al.*, 1993]. While there is some uncertainty in this estimate, it implies that export of NO_x from surface sources is about 3 times larger than the aircraft source, which is 2% of the surface source. *Jacob et al.* [1993] estimated that the amount of ozone produced by the NO_x that is exported is about equal to the amount of ozone produced in the boundary layer from surface emissions and subsequently exported. *Ehhalt et al.* [1992], using a simple quasi-2-D model, estimated that aircraft provide 30-40% of the NO_x in the upper troposphere at midlatitudes, with most of the balance provided by surface emissions from eastern North America and Europe that are transported to high altitudes by convection. *Kasibhatla* [1993], using a 3-D model, concluded also that aircraft contribute about 30-40% of NO_x in the upper troposphere at midlatitudes, but that the other major source in this region is the stratosphere. Quantification of the effect of aircraft emissions on ozone requires clarification of the mechanisms controlling the distribution of NO_x in the middle and upper troposphere.

Trends in ozone in the boundary layer are most likely to be affected by trends in surface emissions on a regional scale (several hundred kilometers), as the lifetime of NO_x is short, about a day in summer. In the middle and upper troposphere the lifetime of NO_x is longer and the lifetime of ozone itself is weeks, so that ozone has more potential to be affected by trends in emissions from more distant regions or by aircraft emissions. Any trend in ozone caused by increases in NO_x is superimposed on interannual variability driven by dynamical factors, such as stratosphere-troposphere exchange. In summer, high ozone values in the boundary layer are associated with slow moving high-pressure systems, high temperatures, and low wind speeds [e.g., *Decker et al.*, 1976; *Guicherit and van Dop*, 1977; *Logan*, 1989; *Seinfeld et al.*, 1991]. In winter, high ozone values are associated with high wind speeds and downward transport of ozone [*Davies et al.*, 1992].

Increases in ozone from the ground to 10 km (300 mbar) are found year round only for Hohenpeissenberg. The pattern of an increase in ozone before 1982, but not since, is consistent with the pattern of surface emissions of NO_x in Western Europe (Figure 21). The increase in ozone in summer, about 30% at 500 mbar from the early 1970s to the late 1980s, is larger than the increase in surface sources of NO_x. Model studies over the United States suggest that the fractional response of the ozone concentration in the boundary layer is about one third of the fractional change in NO_x emissions [*McKeen et al.*, 1991; *Jacob et al.*, 1993], but no such estimates are available from global models. Aircraft emissions could have contributed to the trend. The largest increase in ozone occurred, however, in the few years when jet fuel use was constant, 1978-1983, while there has been no increase in ozone since 1982, although aircraft emissions have increased significantly (Figure 22).

Figure 23 illustrates the difficulty of relating increases in aircraft emissions and in ozone in the upper troposphere. Mean summer values of ozone at Hohenpeissenberg are 65-120 ppb at 300 mbar and 80-180 ppb at 250 mbar, while changes in ozone caused by aircraft are estimated to be a few parts per billion over 20 years [*Beck et al.*, 1992; *Johnson et al.*, 1992]. The interannual varia-

Table 10. Trend in Column Ozone Caused by Trends in the Troposphere

Station	Years	Pressure Levels, mbar	%/Decade December-March	%/Decade May-August
Resolute	1980-1991	1000-350	-1.1	NS
Churchill	1980-1991	1000-350	NS	-0.5
Goose Bay	1980-1991	1000-350	-0.7	NS
Wallops Is.	1970-1991	1000-225	NS	1.0
Lindenberg	1975-1991	1000-450	0.8	NS
Hohenpeissenberg	1970-1991	900-275	1.5	1.5
	1980-1991	900-275	0.8	1.3
Sapporo	1970-1991	1000-550	0.5	ND
Tateno	1970-1991	1000-550	0.3	ND
Kagoshima	1970-1991	900-450	0.5	ND
Aspendale	1970-1991	1000-225	-1.4	NS

The last two columns give the tropospheric column trend, expressed as a percentage of the total ozone column. The tropospheric trend was derived by integrating over the pressure levels given in the third column, for the time period given in the second column; the ozone column was obtained by integrating the mean ozone profile defined by the sonde measurements over the same time period. NS indicates that the tropospheric trends were not significant; ND indicates no data.

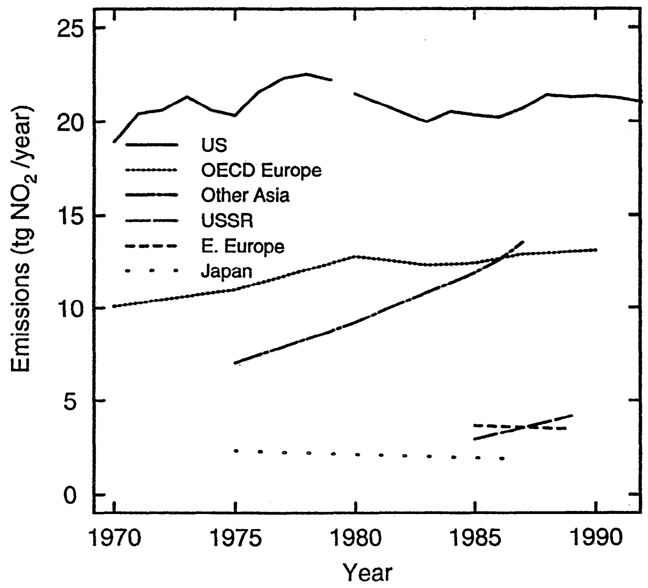


Figure 21. Trend in emissions of NO$_x$ in different regions. Data were taken from *EPA* [1993] for the United States, *OECD* [1993] for Europe, *World Resources Institute* [1993] for Eastern Europe and the former USSR, and *Kato and Akimoto* [1993] for Asia; "Other Asia" refers to Asia, excluding Japan. The break at 1979/80 for the United States indicates that different methodologies were used for the emissions estimates before 1979 and after 1980 [*EPA*, 1993].

tions of ozone at 300 mbar are often, but not always, related to variations at levels up to 125 mbar, indicating a strong dynamical influence, but show no relationship to values at 500 mbar. Ozone values at 300 mbar did not increase during the 1970s, but were much higher in the 1980s than in the 1970s, giving rise to the trend for 1970-1991. The difference in ozone between the 1980s and 1970s could simply be a dynamical phenomenon, reflecting a larger influence of air from the lower stratosphere in the 1980s. In 1991-1993, ozone at 300 mbar dropped down to values seen in the 1970s, while ozone at 500 mbar and below remained 5-10 ppb higher than values in the early 1970s.

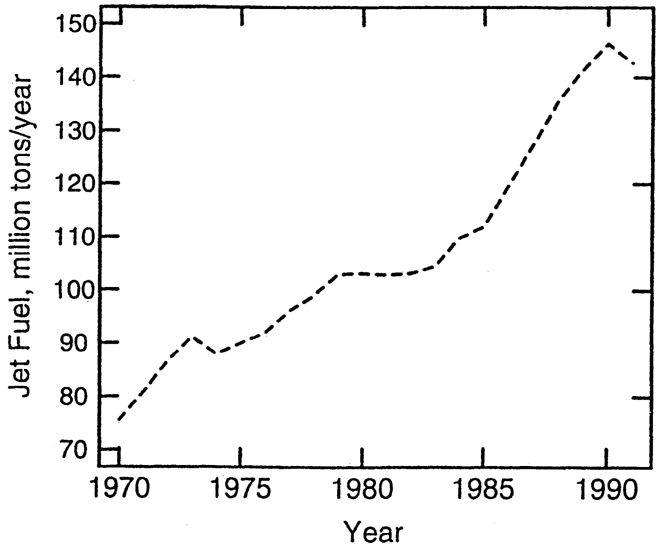


Figure 22. Trend in use of jet fuel. Statistics for worldwide consumption were taken from *United Nations* [1993]; they are the sum of the "consumption" and "bunkers" categories.

The smallest trends in ozone among the developed nations are those over North America. Wallops Island shows a small trend in summer, ~1% yr^{-1} from 300 mbar to the ground, for 1970-1991, and a smaller insignificant trend for 1970-1992. Much of the trend is caused by low values in the early 1970s. There appears to be no increase in ozone in the middle troposphere over Boulder, upwind of the eastern United States. Urban areas of the United States show no trend in ozone in the 1980s [*EPA*, 1992b]. Trends in nonurban ozone are not to be expected if surface emissions are the largest anthropogenic source of NO$_x$ over the United States. There is no evidence that aircraft emissions have affected ozone. At Wallops, as at Hohenpeissenberg, variations in ozone at 300 mbar are related to variations up to about 125 mbar but not to variations at 500 mbar.

None of the Canadian stations show a long-term increase in ozone, and three show a decrease since 1980. The recent decrease is significant only in winter (Resolute and Goose Bay) and autumn (Resolute and Churchill) and may reflect dynamical variability and lower amounts of stratospheric ozone. Surface data from Barrow, Alaska, give an annual increase of 0.7% yr^{-1} since 1973, significant only in summer when the trend is 1.7% yr^{-1}; the site may be influenced by emissions from the Alaskan oil industry [*Oltmans and Levy*, 1994]. Analysis of aircraft data in the regions around Churchill and Goose Bay showed that ozone near 500

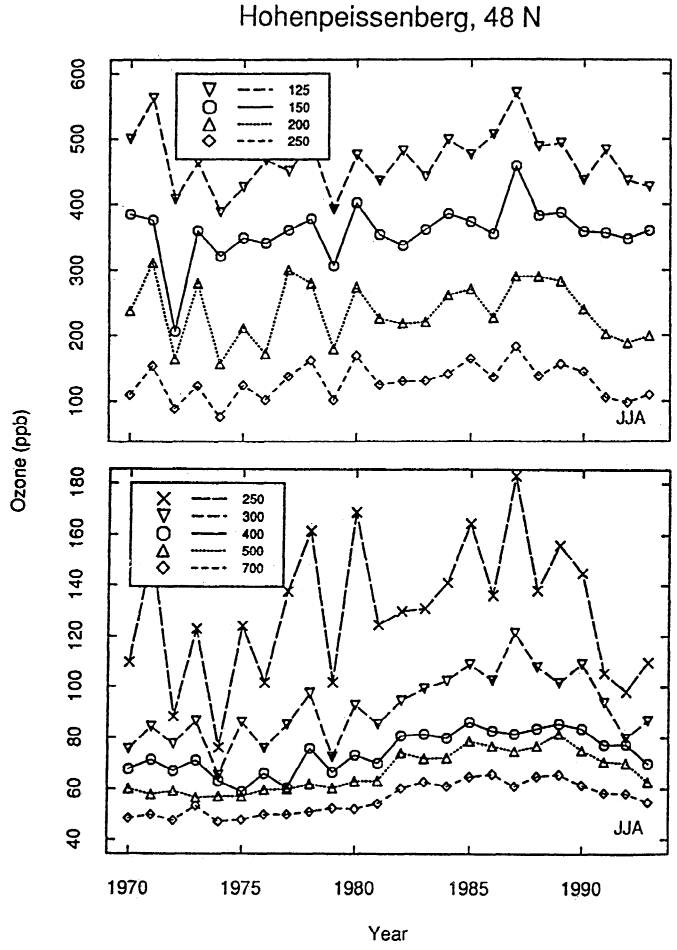


Figure 23a. Mean summer concentrations of ozone at Hohenpeissenberg for (bottom) 700-250 mbar and (top) 250-125 mbar 1970 to 1992. Note the different scales for the two panels.

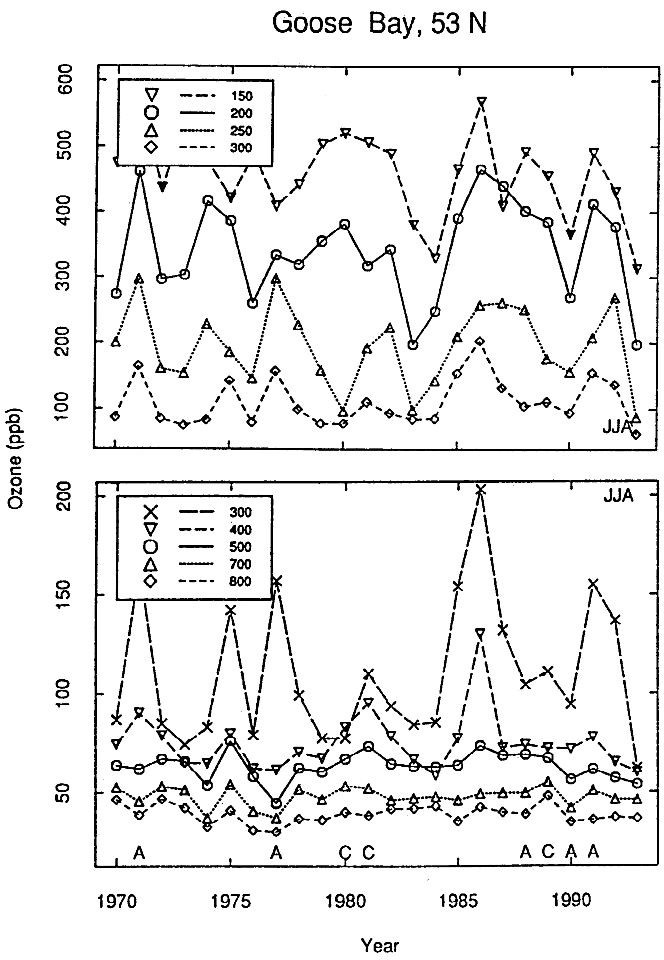


Figure 23b. Mean summer concentrations of ozone at Goose Bay for (bottom) 800-300 mbar and (top) 300-150 mbar for 1970 to 1992. Note the different scales for the two panels. The letters "C" show the years for which the area of fires in Canada exceeded twice the mean area, and the letters "A" show the same information for Alaskan fires.

mbar is affected by aged stratospheric intrusions about 40% of the time in late summer, as diagnosed from ozone and aerosol measurements [*Browell et al.*, 1994] and analyses of potential vorticity [*Bachmeier et al.*, 1994]. Forest fires appear to provide the largest source of NO_x to this region in summer, rather than transport of NO_x from fossil fuel use in the United States and southern Canada [*Fan et al.*, 1994]. However, the years with the largest forest fires in Canada and Alaska are not the years with highest ozone, as shown in Figure 23b. Goose Bay is near the trans-Atlantic air corridor. Ozone is extremely variable in the upper troposphere in summer, with mean values of 60-130 ppb at 400 mbar and 75-200 ppb at 300 mbar (Figure 23b). At Goose Bay, the interannual variability at 500 mbar bears some resemblance to that at 300-150 mbar, suggesting more influence of lower stratospheric air here than at lower latitude locations.

Japan is downwind of China, where surface emissions of NO_x are steadily increasing [*Kato and Akimoto*, 1992]. The trend in ozone below 5.5 km (500 mbar) over Japan may well reflect this increase [*Akimoto et al.*, 1993]. The large trends near the surface are most likely to reflect local trends in SO_2 and NO_x. There has been a small increase in ozone at Mauna Loa, Hawaii, 0.37% yr^{-1} since 1973 [*Oltmans and Levy*, 1994]. The site is downwind of Asia, and export of pollution is expected to be strongest in winter and spring [*Balkanski et al.*, 1992]. The trend is largest in winter and spring but is not significant in either season. There does not appear to be any trend in ozone at Mauna Loa since 1980.

Large increases in surface emissions of NO_x are not expected in developed nations over the next decade because of increasing implementation of emission controls on vehicles and power plants and slow growth of fossil fuels [e.g., *EPA*, 1993]. Significant increases are expected in China, where consumption of fossil fuels increased by 5% yr^{-1} in the 1980s, and where there are few environmental controls. Emissions from subsonic aircraft are projected to increase by 85% from 1990 to 2015 [*Wuebbles et al.*, 1993].

Ozonesondes will continue to provide the only information on trends in ozone in the troposphere and lower stratosphere in the coming decade, with the exception of a few lidar stations. Sondes have the potential to provide high-quality data, but improvements in quality are required for many stations. In order to determine tropospheric trends in the future, the measurement frequency should be be increased to a minimum of weekly, and the spatial coverage should be improved, particularly in the tropics and subtropics.

5. Conclusions

Trends in tropospheric ozone are highly variable. There is no long-term change in ozone over Canada, the region of the northern latitudes least likely to be affected by sources of NO_x from fossil fuel combustion. Ozone has increased slightly in summer over the east coast of the United States, but the significance of the increase disappears with the inclusion of one additional year of data; data from Boulder show no evidence for a trend in the middle and upper troposphere. The Japanese stations show increases in ozone below 5.5 km, likely caused by increases in surface emissions on the Asian continent. Large increases in ozone (2% yr^{-1}) are found year round only for Hohenpeissenberg and Payerne, and the Payerne record is presently under revision. The increase at Hohenpeissenberg leveled off in the 1980s, and values have been decreasing since 1989. The reason for higher ozone trends over these European stations than over the eastern United States is unclear, since the trends in surface emissions of NO_x have been similar. Aircraft emissions could have contributed to the ozone increases. The lack of an increase in ozone in the last decade, during a period of rapid growth of aircraft emissions, argues against a significant influence. The higher ozone values in the upper troposphere in the 1980s compared with the 1970s over Europe could be caused simply by dynamical factors.

To gain further insights into mechanisms responsible for trends in tropospheric ozone will require improved understanding of (1) the factors that control NO_x concentrations in the middle and upper troposphere and (2) the dynamical factors that influence the interannual variability of ozone. Analyses of the effects of meteorology and dynamics on ozone on a year-by-year basis for the regions discussed here would aid considerably in deconvolving the effects of trends in precursor emissions; a similar recommendation was made recently by *Beekman et al.* [1994].

Decreases in stratospheric ozone are found from about 24 km to near the tropopause; the decreases near 17 km are smaller than those derived from SAGE for midlatitudes. Decreases in ozone below 17 km are large enough to account for the 20% difference between column ozone trends derived from TOMS [*Stolarski et al.*, 1992] and SAGE [*McCormick et al.*, 1992]. The ozone increases found over Hohenpeissenberg make a significant contribution to the trend in the ozone column. The seasonal losses in

Table A1. Serial Correlation in the Ozone Sonde Data

	Hohenpeissenberg				Payerne			
Pressure, mbar	ϕ_1	ϕ_2	ϕ_3	ϕ_4	ϕ_1	ϕ_2	ϕ_3	ϕ_4
50	0.30	-0.03	0.17		0.17	0.05	0.08	0.14
80	0.05	0.22	0.16		0.11	0.09	0.18	
500	0.16	0.08	0.14	0.27	0.44	0.18	-0.01	0.18

The table gives the coefficients ϕ_i in equation (A2). The analysis used data from 1970 to 1991.

ozone in different parts of the world are consistent with results for the ozone column shown by *Stolarski et al.* [1992], *Niu et al.* [1992] and *Reinsel et al.* [1994]. Ozone is decreasing year round on the edge of the Antarctic continent.

Trends derived from the sonde data are dependent on ozone column measurements. While potential errors in the column data affect the exact magnitude of trends presented here, they do not affect the major conclusions of this study. Corrections to the historical column data should be incorporated into the sonde records and included in future studies.

Appendix: Temporal Correlation in the Ozonesonde Data

Models 1 and 2 (section 2.2) may underestimate the error in the ozone trend, because they do not account for temporal correlation in the ozone time series [*Tiao et al.*, 1990; *WMO*, 1990a, Appendix A]. I explored this issue, using a model similar to the ones adopted by *Bojkov et al.* [1990], and *WMO* [1992]:

$$y(t) = = \sum_{i=1}^{x} a_i \, I_{i;t} t + b\mathrm{QBO}(t-k) + c\mathrm{SOL}(t) + N(t). \qquad \text{(A1)}$$

where $I_{i;t}$ is an indicator series for the *i*th month of the year, 1 if the month *t* corresponds to month *i* and zero otherwise, and a_i is the trend for month *i*. In model 3 the 12 monthly trends, *b*, and *c* are determined simultaneously by using a generalized linear model. The same dependence on the QBO and and solar flux functions is assumed implicitly year round. Temporal correlation in the ozone values was examined by allowing $N(t)$ to be an autoregressive series,

$$N(t) = \phi_1 N(t-1) + \phi_2 N(t-2) + \ldots + e(t), \qquad \text{(A2)}$$

where $e(t)$ is an uncorrelated series, and by determining the order of $N(t)$ and the values of ϕ_i.

Model 3 was applied to observations from Hohenpeissenberg and Payerne, the stations with the highest measurement frequency. The coefficients ϕ_i in the autoregressive series $N(t)$ are given in Table A1. In the region of peak ozone loss in the stratosphere, $N(t)$ is of the order of 3, and for the mid-troposphere, $N(t)$ is of the order of 4. Analyses of trends in the ozone column have assumed $N(t)$ to be of the order of 1 [*Bojkov et al.*, 1990; *WMO*, 1992; *Bjarnason et al.*, 1993]. Inclusion of temporal correlation in the analysis should make little difference to the slope of the regression line but should give a more realistic, and larger, standard error [*Tiao et al.*, 1990; *WMO*, 1990a, Appendix A]; for $N(t)$ of the order of 1, the standard error is larger than the standard error in the model that does not account for temporal correlation by the factor

$$((1 + \phi)(1 - \phi))^{0.5}.$$

If only the first-order terms given in Table A1 are considered, for ϕ in the range 0.17 to 0.44, this factor is 1.2 to 1.6.

It does not appear, on the basis of the results in Table A1, that the underestimate in computing the standard errors with models 1 and 2 is serious enough to invalidate the conclusions presented here. The sonde data are not well suited for application of model 3, developed for ozone column data. There are substantial gaps in the record for about half the stations. The sonde data are noisier than the column data, in part because of the lower sampling frequency. Many of the monthly trends are not significant, even when the seasonal trends are significant, a consequence of the small sample size.

Acknowledgments. I would like to thank J. Staehelin for providing the Payerne data, and for many communications about the data. I would also like to thank V. Kirchhoff, S. Oltmans, and A. Torres for providing data that were not available from WODC, and L. Morrison and E. Hare for providing the data from WODC. D. Jacob and C. Spivakovsky provided useful comments on this paper. This work was supported by NSF grant ATM-8921119 and DOE grant DE-FG02-93ER61709 to Harvard University.

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(Received April 28, 1994; revised August 24, 1994;
accepted August 30, 1994.)