JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 104, NO. D13, PAGES 16,115–16,149, JULY 20, 1999

An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D models and development of a gridded climatology for tropospheric ozone

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Abstract. I present an analysis of ozonesonde data, synthesizing what is known about the distribution of tropospheric ozone. Major features of the distribution are highlighted, and recommendations are given for testing three-dimensional models of tropospheric chemistry and transport with these data. The data are analyzed on pressure surfaces and relative to the height of the thermal tropopause. A minimum of 20 soundings are required for 95% confidence intervals of the ozone monthly means to be less than ±30% near the extratropical tropopause. Twenty soundings also ensures means reliable to better than ±15% for 800-500 hPa for the extratropics and for 800-100 hPa in the tropics. Ozone variability is higher in the upper troposphere for subtropical locations than for tropical locations, and 35 soundings are required for 400-100 hPa for the means to be defined to better than ±15%. For northern middle and high latitudes, the broad summer maximum in ozone in the middle troposphere extends all the way up to the tropopause. Median concentrations at the tropopause are highest in June and July, typically 125-200 ppb, and are a factor of 2 smaller in winter. Highest values of ozone are in spring 2 km above the tropopause. The change in the phase of the annual cycle of ozone between the tropopause and the region immediately above it, and the steep concentration gradients across the tropopause, suggest that high vertical resolution (~1 km) will be required in models to simulate this behavior. Mean ozone values in the middle troposphere are approximately constant from 30° to 75° in the winter in both hemispheres, while there is a maximum from 35° to 50°N in summer. In the northern subtropics, there is a summer minimum in middle tropospheric ozone over the Pacific and a summer maximum over the Atlantic which appear to be related to differences in circulation. Mean ozone values over Samoa are similar to those measured 20-30 years ago over Panama. Ozone is higher over the tropical South Atlantic (Natal) than over the western Pacific (Samoa) all year from about 800 hPa to the tropopause; ozone is most similar in May and June over the Atlantic and Pacific, the months with minimum burning in the tropics. The ozone maximum at Samoa in the middle and upper troposphere in October is caused by long-range transport of ozone and its precursors from biomass burning, with the peak lagging that at Natal by about a month. The secondary peak in ozone in January and December at South Atlantic sites reflects transport of biomass burning effluents from the Northern Hemisphere. The sonde data were used in combination with surface and satellite data to derive a gridded climatology for tropospheric ozone.

1. Introduction

Ozone plays an important role in controlling the chemical composition of the troposphere. Photolysis of ozone near 300 nm,

$$O_3 + h\nu \rightarrow O(^1D) + O_2$$

followed by reaction of $O(^1D)$ with water,

$$O(^1D) + H_2O \rightarrow OH + OH$$

leads to production of OH. Reaction with the OH radical is the primary removal process for a variety of trace gases of importance to tropospheric and stratospheric chemistry, including CO, CH_4, NO_2, and halocarbons. Ozone also 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]. Increases in tropospheric ozone cause an increase in surface temperature, with the climate forcing most sensitive to ozone changes in the middle troposphere in a model with cloud feedback [*Hansen et al.*, 1997]. High concentrations of ozone in the lower troposphere are of concern because of the deleterious effect on human health and vegetation [e.g., *Horvath and McKee*, 1994]. Ozone is thought to be responsible for most of the crop damage caused by air pollution in the United States [e.g., *Heck et al.*, 1983; *Tingey et al.*, 1994] and may be contributing to observed damage to forests in Europe and the eastern United States [*McLaughlin and Downing*, 1995; *Skarby et al.*, 1995].

Ozone enters the troposphere from the stratosphere and is removed at the surface of the earth by vegetation [*Danielson*, 1968; *Galbally and Roy*, 1980]. Ozone is also formed in the troposphere during the oxidation of CO, CH_4, and hydrocarbons in the presence of NO_x, and destroyed by reactions with HO_x radicals

Paper number 1998JD100096.
0148-0227/99/1998JD100096$09.00

[e.g., *Crutzen,* 1973; *Chameides and Walker,* 1973; *Logan,* 1985; *Warneck,* 1988]. The photochemical source of ozone is critically dependent on the amount of NO_x in the atmosphere. A variety of modeling approaches have been used to quantify the sources and sinks of ozone. Detailed chemistry models, constrained by observed concentrations of trace gases, have been used to evaluate local production and loss rates [e.g., *Chameides et al.,* 1987, 1989; *Carroll et al.,* 1990; *Davis et al.,* 1996; *Jacob et al.,* 1996]. In recent years three-dimensional (3-D) chemical transport models, with varying levels of sophistication in the representation of ozone chemistry, have become the primary tool for investigating the mechanisms controlling ozone and for quantifying its sources and sinks [*Levy et al.,* 1985; *Crutzen and Zimmerman,* 1991; *Muller and Brasseur,* 1995; *Roelofs and Lelieveld,* 1995; *Roelofs et al.,* 1997; *Kasibhatla et al.,* 1996; *Levy et al.,* 1997; *Berntsen and Isaksen,* 1997; *Houweling et al.,* 1998; *Wang et al.,* 1998a, b, c]. Photochemical production of ozone in these models is ~3500-4000 tg yr^{-1}, photochemical loss ~3000-4000 tg yr^{-1}, stratospheric input ~400-850 tg yr^{-1}, and surface removal ~500-1200 tg yr^{-1}. The 3-D models are also being used to assess the impact of subsonic aircraft on ozone and other trace gases in the tropopause region [e.g., *Kasibhatla,* 1993; *Brasseur et al.,* 1996; *Flatoy and Hov,* 1996; *Kraus et al.,* 1996; *Friedl,* 1997].

An essential step in the development and application of models for atmospheric chemistry and dynamics is their evaluation with observations [*Prather and Remsberg,* 1993; *Stolarski et al.,* 1995]. After model development, model evaluation is often the most demanding task, because of the significant effort involved in collecting and analyzing the data. A major goal of this paper is to highlight the key features of the ozone distribution that a credible model should be able to reproduce and to provide the data to the modeling community. The second goal of the paper is to provide a gridded climatology of tropospheric ozone based on sonde, surface, and satellite data for ozone. This climatology may be used to derive empirically based estimates of chemical sources and sinks for ozone and to calculate the global distribution of OH [e.g., *Spivakovsky et al.,* 1990]. It may also be used to provide a priori profiles for satellite retrievals, for climate studies, and initial conditions for 3-D chemical transport models. The third goal of this paper is to emphasize regions where measurements are most needed to improve current understanding of tropospheric ozone.

The data used here are primarily measurements of the vertical distribution of ozone using ozonesondes, although use is made also of surface measurements. Vertical profiles of ozone are available from aircraft surveys, but these provide "snapshots" of the ozone distribution for short time periods [e.g., *Marenco et al.,* 1989, 1990; *Browell et al.,* 1996]. My objective is to provide long-term averages that can be used to evaluate 3-D models driven either by winds from a general circulation model or by mean winds derived from observations. Models driven by assimilated winds for specific periods are best evaluated with contemporaneous data [e.g., *Rood et al.,* 1991; *Douglass et al.,* 1997; *Law et al.,* 1998]. I present the ozone analysis using pressure as a vertical coordinate, and I show also the ozone data on a vertical scale relative to the tropopause height. The latter approach emphasizes the different behavior of tropospheric versus stratospheric ozone near the tropopause, a region often not well represented in low-resolution 3-D models. An analysis of the sonde data with a focus on the stratosphere is given in a companion paper [*Logan,* this issue].

Measurements of ozone have been made since 1994 on five commercial aircraft as part of the MOZAIC program [*Marenco et al.,* 1998]. The first two years of these data were used to develop mean distributions of ozone in the upper troposphere (8-12 km) over much of northern midlatitudes and are also being used to evaluate 3-D models [*Thouret et al.,* 1998a; *Law et al.,* 1998]. The program also provides vertical distributions of ozone near airports, and these data were compared to the sonde data shown here for several locations and found to be in good agreement [*Thouret et al.,* 1998b]. The data from MOZAIC complement the sonde data, in that they provide better spatial coverage in the upper troposphere and they provide vertical profiles for some locations in the tropics and Southern Hemisphere where sonde data are lacking [*Thouret,* 1998]. Lidar measurements of tropospheric ozone are also being made at a few locations, in Europe and elsewhere [*Beekmann et al.,* 1994a; *Hov,* 1997; *WMO,* 1998]. The measurements from MOZAIC and from lidars were not used in this study but offer great potential for improving knowledge of the ozone climatology in the future.

Some features of the global distribution of tropospheric ozone have been known for over 20 years, based on measurements from the North American Ozonesonde Network which operated in the 1960s [*Hering and Borden,* 1967], and from a few stations in North America, Europe, and Australia, which were established in the late 1960s [e.g., *Dutsch,* 1966; *Dutsch et al.,* 1969; *Pittock,* 1977]. It was noted that concentrations of ozone were higher at midlatitudes of the Northern Hemisphere than in the northern tropics and were higher in the Northern Hemisphere than in the south [*Pruchniewicz,* 1973; *Chatfield and Harrison,* 1977a; *Pittock et al.,* 1977; *Fishman et al.,* 1979b]. Ozone values at northern midlatitudes were highest in spring and summer in the middle troposphere [*Chatfield and Harrison,* 1977a; *Fishman et al.,* 1979b]. Subsequent measurements supported these conclusions, with the exception of the hemispheric gradient in the tropics. The early analysis [*Fishman et al.,* 1979b] used measurements from the southern tropics made with Regener sondes whose use was discontinued in 1965; the old sondes significantly underestimated values for tropospheric ozone [*Chatfield and Harrison,* 1977a, b]. These old data continue to appear in current publications [e.g., *Andreae et al.,* 1992; *Lindesay et al.,* 1996] and should be replaced by more reliable recent data. Measurements of ozone from Natal, Brazil (6°S, 35°W), started in 1979, showed ozone values significantly higher than those reported earlier for the northern tropics; concentrations over Natal peak in austral spring and are as high as values at northern midlatitudes in summer [*Logan,* 1985; *Logan and Kirchhoff,* 1986]. Subsequently, data from Samoa (14°S, 170°W) [Oltmans et al., 1989a] showed evidence for considerable longitudinal gradients in tropical ozone, with lower values over Samoa than Natal but similar seasonal cycles in the middle troposphere.

Logan [1985] analyzed data from about 30 stations and noted a broad summer maximum in the lower troposphere (700 hPa) over Europe, the United States, and northern Japan but a late spring maximum over Canada. Ozone concentrations in summer over the United States and Europe appeared to exceed those over Canada by 5-15 ppb. The summer maximum at 500 hPa extends as far south as 21°N in the western hemisphere but not over Japan, where ozone decreases dramatically in early summer at 32°N, because of inflow of tropical marine air. Highest concentrations of ozone in the lower and middle troposphere of the Northern Hemisphere were found between 30° and 50°. *Oltmans et al.* [1989a] showed that ozone concentrations over New Zealand (45°S) are higher than those over Samoa and the Antarctic continent all year except for austral spring, when similar values are found over New Zealand and Samoa.

Fishman et al. [1990] presented the first global picture of the

Table 1. Ozonesonde Data

WMO Code	Station	Latitude	Longitude	Altitude, m	Type	Number	Data Record
18	Alert	82°N	62°W	62	ECC	347	1/88-12/93
89	Ny Alesund	79°N	12°E	0	ECC	231	10/90-12/93
24	Resolute	75°N	95°W	64	ECC	540	1/80-12/93
262	Sodankyla	67°N	27°E	179	ECC	246	1/89-12/92
77	Churchill	59°N	147°W	35	ECC	515	1/80-12/93
21	Edmonton	53°N	114°W	668	ECC	498	1/80-12/93
76	Goose Bay	53°N	60°W	44	ECC	546	9/80-12/93
221	Legionowo	53°N	21°E	96	GDR[a]	234	1/80-12/93
174	Lindenberg	52°N	14°E	98	GDR[a]	845	1/80-12/93
174	Lindenberg	52°N	14°E	98	ECC	212	6/92-12/95
99	Hohenpeissenberg	48°N	11°E	975	BM	1615	1/80-12/93
156	Payerne	47°N	7°E	491	BM	1142	1/80-12/89
197	Biscarosse	44°N	1°W	18	BM	338	3/76-1/83
132	Sofia	43°N	23°E	588	GDR	195	2/82-12/91
12	Sapporo	43°N	141°E	19	KC	250	1/80-12/95
67	Boulder[b]	40°N	105°W	1634	ECC	333	1/85-12/93
38	Cagliari	39°N	9°E	4	BM	302	7/68-7/80
107	Wallops Is.[b]	38°N	76°W	4	ECC	401	1/80-12/93
14	Tateno	36°N	140°E	31	KC	444	1/80-12/95
7	Kagoshima	32°N	131°E	283	KC	227	1/80-12/95
210	Palestine	32°N	96°W	121	ECC	187	2/75-6/85
	Kennedy[c]	29°N	31°W		BM	147	2/66-2/69
10	New Delhi	29°N	77°E	220	Ind	60	1/69-9/86
190	Naha	26°N	128°E	27	KC	185	9/89-12/95
	Grand Turk[c]	21°N	71°W		BM	80	3/66-5/69
109	Hilo[b]	20°N	155°W	11	ECC	359	1/85-12/93
187	Poona	19°N	74°E	559	Ind	133	2/66-11/86
	Panama[c]	9°N	80°W	57	BM	43	6/66-5/69
203	Panama	9°N	80°W	57	ECC	58	7/77
205	Trivandrum	8°N	77°E	60	Ind	48	7/69-10/86
329	Brazzaville[d]	4°S	15°E	314	ECC	81	4/90-10/92
219	Natal[e]	6°S	35°W	32	ECC	281	11/78-10/92
328	Ascension Is.[d]	8°S	14°W	91	ECC	67	7/90-10/92
191	Samoa[b]	14°S	170°W	82	ECC	164	4/86-10/96
265	Pretoria (Irene)	26°S	28°E	1369	ECC	133	7/90-10/93
26	Asp./Laverton	38°S	145°E	21	BM/ECC	285	1/80-12/95
256	Lauder[f]	45°S	170°E	370	ECC	286	8/86-12/90
233	Marambio	64°S	57°W	198	ECC	241	11/88-12/95
101	Syowa	69°S	39°E	21	KC	303	1/86-12/95

Read 1/88-12/83 as January 1988 to December 1993. The data were obtained from the World Ozone and Ultraviolet Data Center, with the exceptions noted below, and were current as of November 1997. Column 8 shows which data were included in the climatology; 1980-1993 was chosen as the period if such data were available. Later data were included as necessary to increase the number of measurements. Column 7 gives the numbers of soundings that met the selection factor criteria: 0.8-1.2 for ECC and KC sondes, 0.9-1.35 for the Brewer Mast type of sondes (BM, Indian, and GDR sondes), except for Hohenpeissenberg, for which 0.9-1.2 was used.

[a]Legionowo changed to ECC sondes in June 1993; Lindenberg changed to ECC sondes in June 1992.

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

[c]Data provided by R. Chatfield, originally collected by *Hering and Borden* [1967].

[d]Data obtained from the NASA/GTE data archive at NASA Langley.

[e]Data provided by V. Kirchhoff.

[f]Data provided by G. Bodeker.

distribution of tropospheric ozone, obtained by subtracting the amount of stratospheric ozone derived from the Stratospheric Aerosol and Gas Experiment (SAGE) from the overhead column of ozone obtained from the Total Ozone Mapping Spectrometer (TOMS). The resulting product, the tropospheric ozone residual, is the column of ozone up to the local tropopause. The residual shows clearly the longitudinal gradient in ozone in the southern tropics, with highest values over the south tropical Atlantic in austral spring and lowest values over the western Pacific near the equator. Highest values at midlatitudes are found downwind of Asia, North America, and Europe in summer.

The sources of data and the analysis procedures used in this study are described in section 2, key features of the ozone distribution are discussed in section 3, the gridded ozone climatology is derived in section 4, recommendations for the use of data to test models are given in section 5, and conclusions and recommendations are given in section 6.

2. Analysis of Ozone Measurements

Data from the ozonesonde stations in Table 1 were considered in this analysis. The primary source for data was the World Ozone and Ultraviolet Data Center (WOUDC), Environment Canada, Downsview, Ontario (http://www.tor.ec.gc.ca/ woudc). Data were also provided by S. Oltmans, J. Staehelin, A. Torres, V. Kirchhoff, and G. Bodeker. The measurements were made

with two types of ozonesonde: the electrochemical concentration cell (ECC) [*Komhyr,* 1969; *Komhyr et al.,* 1995] and variations on the Brewer Mast (BM) bubbler [*Brewer and Milford,* 1960; *Beekmann et al.,* 1994a]. Both types are based on the reaction of potassium iodide (KI) with ozone. The sonde techniques and associated errors are discussed in detail in *WMO* [1998]. Data quality issues are also discussed by *Reid et al.* [1996], *WMO* [1995], *Logan* [1985, 1994], *Tiao et al.* [1986], and references therein.

Brewer Mast sondes underestimate stratospheric ozone. It is standard practice for all types of sonde measurements to integrate the vertical profile of ozone, allowing for the ozone above the altitude reached by the sonde, and to scale the profile to a concurrent Dobson or Brewer measurement of the ozone column. Most of the sonde measurements used here have 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 [*Logan,* 1985, 1994]. There are 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. The correction factors are used as a quality check; they depend primarily on the reliability of the measurement of stratospheric ozone. The selection criteria adopted here (see Table 1) are discussed by *Logan* [1994].

Four intercomparisons held from 1970 to 1989 found that ECC sondes measured more ozone in the midtroposphere than BM sondes by about 15%-20%; 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; *Beekmann et al.,* 1994a]. An intercomparison in 1991 gave opposite results, with BM sondes giving tropospheric values 10%-25% higher than ECC sondes [*WMO,* 1991]. Assessments of the accuracy of ozonesondes suggest that ECC sondes overestimate tropospheric ozone by about 5%, while BM sondes underestimate tropospheric ozone by about 9% [*Komhyr et al.,* 1995; *Beekmann et al.,* 1994a; *Reid et al.,* 1996], consistent with the results of the earlier intercomparisons. However, a recent comparison of ECC sondes from several laboratories with an accurate UV photometer in an environmental chamber shows that the bias may be positive or negative, depending on the laboratory, but is less than ±5% [*Smit et al.,* 1998]. The same study showed that the precision of ECC sondes, ±5%, appears to be higher than those of the BM, Japanese and Indian sondes, ±10-15%. Errors are largest in the upper troposphere where absolute concentrations of ozone are smallest. Most of the ozone stations in the Southern Hemisphere and in northern middle and high latitudes use ECC sonde types (ECC and KC in Table 1), while most stations in the northern tropics and subtropics used Brewer type instruments (BM and Indian in Table 1). No attempt was made here to adjust for differences in the response of the various sonde types to tropospheric ozone.

Four stations in eastern Europe and one in Antarctica used Brewer sondes manufactured in the former East Germany (type GDR in Table 1) until replaced with ECC sondes in 1992. The GDR sondes appear to give values for tropospheric ozone which are higher than measured with either BM or ECC sondes, as shown by data obtained at Berlin (BM) and nearby Lindenberg (GDR, then ECC) [*Logan,* 1994]. Tropospheric values recorded at Lindenberg with GDR sondes were about 30% higher than values at Berlin and other European stations in the 1970s [*Logan,* 1985]. Values obtained with GDR sondes in Antarctica are about 10 ppb higher than values obtained with ECC sondes. Given the systematic errors apparent in the GDR measurements of tropospheric ozone, I used the GDR data only for studies of the stratosphere. Fortunately, there are alternative sonde stations in Europe and Antarctica.

Brewer Mast sondes manufactured in India (Ind. in Table 1) also gave values that were higher that ECC sondes by about 10-22% at the 1970 intercomparison [*Attmanspacher and Dutsch,* 1970]; the sondes did not perform well at the next intercomparison [*Attmanspacher and Dutsch,* 1981]. Most of the Indian data in the WOUDC archive are from 1966 to 1973, with less than

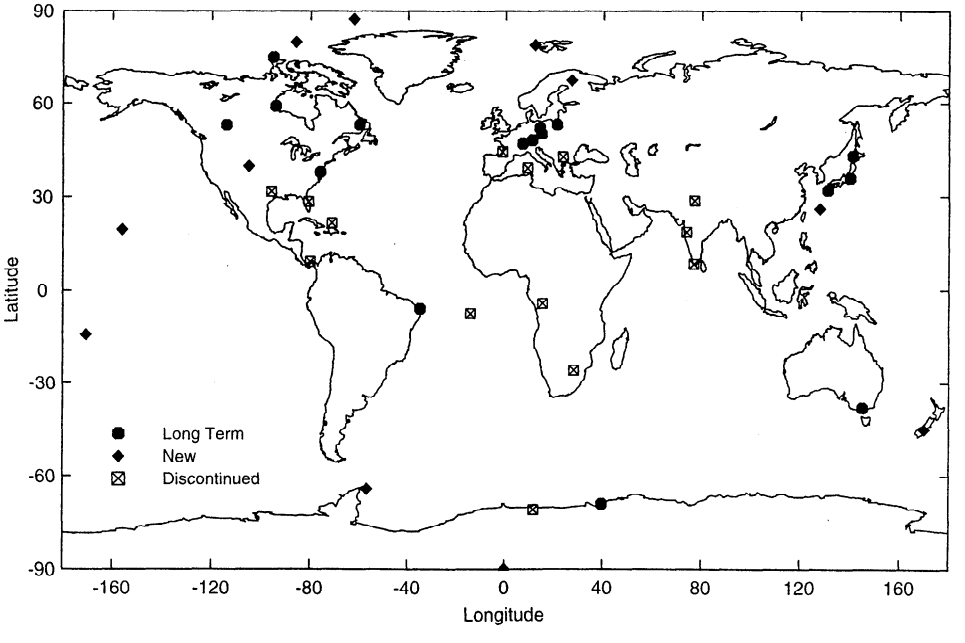


Figure 1. Location of the ozonesonde stations considered in this study. The solid circles show stations that have been making measurements from earlier than 1980; the diamonds show stations that started operation after 1980; and the squares show stations that no longer make measurements (or no longer submit them to WOUDC).

Table 2. Number of Ozone Soundings per Month at 500 hPa

Station	Latitude	J	F	M	A	M	J	J	A	S	O	N	D
Alert	82°N	54	50	44	20	21	20	20	21	22	24	20	31
Ny Alesund	79°N	25	18	30	14	19	12	16	14	15	14	19	35
Resolute	75°N	56	44	47	47	47	44	45	40	38	46	41	45
Sodankyla	67°N	36	30	24	16	17	15	16	18	16	18	17	23
Churchill	59°N	43	48	50	40	42	47	50	45	48	38	30	34
Edmonton	53°N	35	47	46	42	47	45	46	39	42	32	38	39
Goose Bay	53°N	47	44	51	51	47	42	41	45	51	40	42	45
Legionowo	53°N	26	20	25	26	23	14	19	16	20	12	17	17
Lindenberg	52°N	109	113	107	121	47	40	46	33	42	42	38	107
Hohenpeissenberg	48°N	169	158	165	153	112	100	109	104	115	106	159	164
Payerne	47°N	96	92	98	96	99	95	95	90	100	94	92	95
Biscarosse	44°N	24	24	33	27	33	32	28	19	33	33	31	20
Sapporo	43°N	20	20	19	23	28	16	13	18	23	26	19	25
Boulder	40°N	25	28	25	31	29	29	26	22	33	29	30	26
Cagliari	39°N	24	33	36	32	27	27	23	13	15	20	23	28
Wallops Island	38°N	33	35	33	32	46	24	31	37	26	36	37	30
Azores	37°N	0	0	0	0	23	25	16	22	0	0	0	0
Tateno	36°N	52	42	40	40	38	20	23	18	37	39	46	49
Kagoshima	32°N	20	21	23	18	19	15	14	9	19	26	21	22
Bermuda	32°N	0	0	0	51	36	0	23	19	0	0	0	0
Palestine	32°N	0	0	7	4	25	39	9	5	32	51	12	0
Kennedy	29°N	7	9	15	10	20	9	12	11	10	11	14	11
Naha	26°N	16	11	14	16	17	12	15	13	16	19	16	20
Grand Turk	21°N	4	5	6	7	9	8	6	9	5	8	6	7
Hilo	20°N	31	30	27	28	28	29	26	36	31	35	28	29
Poona	19°N	9	9	8	11	5	6	13	10	16	16	17	13
Panama	9°N	2	1	6	4	4	8	2	2	5	4	2	3
Panama	9°N	0	0	0	0	0	0	58	0	0	0	0	0
Brazzaville	4°S	4	5	3	5	4	5	5	11	10	16	8	5
Natal	6°S	23	18	20	23	17	15	29	29	32	31	27	17
Ascension	8°S	6	5	5	7	4	0	2	5	11	14	2	6
Porto Nacional	11°S	0	0	0	0	0	0	0	0	12	0	0	0
Samoa	14°S	13	9	9	12	16	13	11	15	16	23	14	13
Cuiaba	15°S	0	0	0	0	0	0	0	0	13	9	0	0
Namibia	19°S	0	0	0	0	0	0	0	0	6	10	0	0
Irene	26°S	10	11	7	7	12	12	7	6	15	23	11	12
Aspendale/Laverton	38°S	12	22	23	23	22	26	27	32	27	28	24	19
Lauder	45°S	18	15	15	17	17	16	17	40	34	38	31	28
Marambio	64°S	12	11	7	4	3	7	19	34	40	45	39	19
Syowa	69°S	18	20	28	21	11	18	15	29	49	37	33	24
Forster	71°S	25	22	18	17	23	29	26	33	35	40	43	28

a quarter of the data from 1983 to 1986. I compare the results from Poona, the station with the most measurements, with other stations in the subtropics but recommend that the Indian stations are not used for model evaluation.

I selected 1980-1993 as the base period of analysis, but not all stations operated during this period. This period coincides with the climatology available for column ozone from the TOMS (total ozone mapping spectrometer) instrument [*McPeters and Labow,* 1996]. Several years are needed to provide adequate statistics for monthly mean values, since many stations make measurements only once a week. Earlier data were included to increase the geographical coverage, especially in the tropics, and later data were included for stations that started after 1980 and for the Japanese stations where summer data are sparse in the 1980s. The locations of the stations are shown in Figure 1.

In forming monthly statistics for ozone, it would be preferable to derive quantities that reflect interannual variability, as is done for other geophysical quantities [*Oort,* 1983; *Shea,* 1986]. This is not practical however, with only 3-5 acceptable soundings a month for many stations, and even fewer soundings per month for most tropical stations. The statistics given here are for pooled data; that is, the January mean is for all measurements made in any January in the given time period. Trends in tropospheric ozone are small, less than ±1% yr^{-1} since 1980 for Northern Hemisphere stations; tropical data for trends are generally lacking [*Logan,* 1994; *WMO,* 1998].

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. Soundings were omitted which contained negative ozone values. Tables were compiled for each station giving for each level the sample monthly mean, the standard deviation (σ, the square root of the sample variance), and the number of soundings (N); the standard error, SE $=\sigma/\sqrt{N}$ is readily calculated. The 1960s data from the North American Ozonesonde Network in the 1960s analyzed by *Chatfield and Harrison* [1977a] were available on an altitude grid for the troposphere only and were interpolated to a pressure grid using standard atmospheres. Table 2 gives the number of acceptable soundings each month for each station. Monthly statistics for all stations are available from http://www-as.harvard.edu.

The 95% confidence intervals for the monthly means (2 SE) clearly depend on both the inherent variability of ozone and the number of measurements. Many sonde stations at middle and high latitudes have sufficient measurements that 2 SE are <10% of

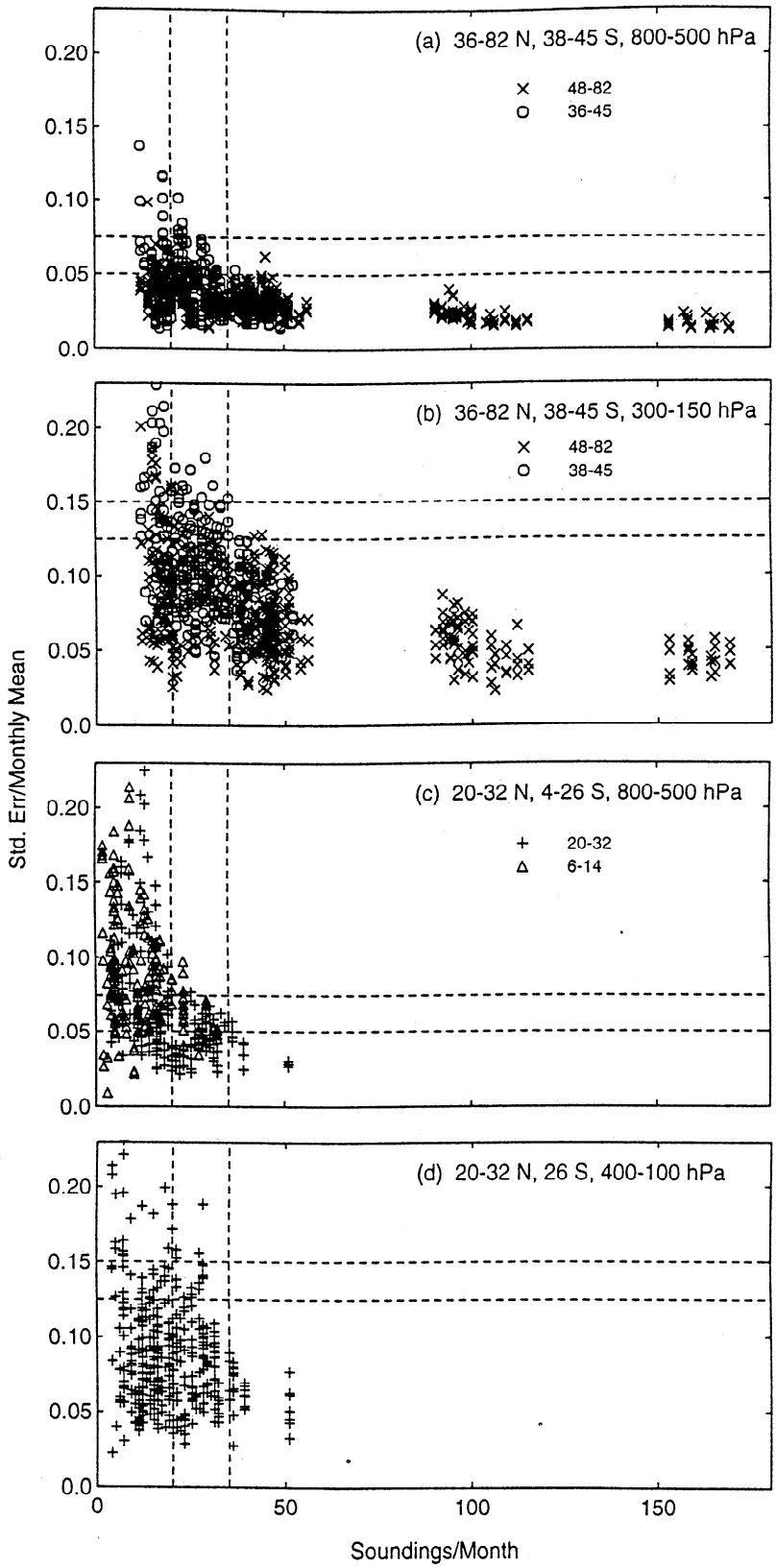


Figure 2. Standard error/monthly mean versus the number of soundings in a month, for the pooled data (see text). The vertical dashed lines are for N=20 and N=35. The horizontal dashed lines are for relative standard errors of 0.05 and 0.075 for Figures 2a and 2c, and for 0.125 and 0.15 for Figures 2b and 2d. Each point is the value for a given station and given month, for one of the pressure levels described in the legend. (a) results for stations at midlatitudes listed in Table 2 for 800-500 hPa; results for the same stations for 300-150 hPa are given in Figure 2b. (c) Results for the subtropics (Kagoshima, Palestine, Naha, Hilo, and Pretoria) and tropics (Brazzaville, Ascension, Natal, and Samoa) for 800-500 hPa and (d) for the subtropics only for 400-100 hPa.

the monthly mean in the middle and lower troposphere. Figure 2a shows that ~20 measurements are required for 1 SE to be <7.5% of the mean, and ~35 measurements for 1 SE to be <5% of the mean even for N<35. (These conclusions apply to measurements made over several years; daily profiles for ozone in a single month are not generally available [*Oltmans et al.,* 1996a]). The variability of ozone is less in winter compared to summer in the middle troposphere. Standard errors are largest near the tropopause (1 SE is usually <15% of the mean for N>20 and <12.5% of the mean for N=35 or more (Figure 2b)); here the ozone distribution is bimodal, rather than approximately normal, as in the troposphere and stratosphere (Figure 3). For most high- latitude stations, 1 SE is <12.5% of the mean near the tropopause even for N<35. In the tropics and subtropics, 20 measurements are required for 1 SE to be <7.5% of the mean below 500 hPa; only two stations have more than 35 soundings in any month (Figure 2c). It appears that there may be higher variability in tropical and subtropical ozone than in midlatitude ozone in the lower troposphere, as the standard errors for the former with 12-20 soundings are larger than those for the latter with the same number of soundings (compare Figures 2c and 2a). For 400-100 hPa the relative standard errors are similar to those in Figure 2c for the tropical stations (Samoa, Natal, Ascension, Brazzaville), while for the subtropical stations (Kagoshima, Palestine, Naha, Hilo, Pretoria), the relative standard errors for 400-100 hPa in Figure 2d are similar to those for higher latitudes (Figure 2b). This is because there is higher variability in the upper troposphere in the subtropics. Variability is highest near the tropopause for these latitudes also.

The standard errors shown for stations with more than 90 soundings may be underestimates, as the inherent assumption that the measurements are independent likely does not hold, since they are made every other day. The high measurement frequency permits a reliable estimate of standard errors for interannual variability for these stations, and for Hohenpeissenberg 1 standard error of the 14 monthly means is typically 2-3% of the overall mean for 800-400 hPa and 4-9% of the overall mean for 400-150 hPa.

The results in Figure 2 show that for monthly mean ozone values to have 95% confidence intervals of ±15% throughout the troposphere, a minimum of 20 soundings is required in the tropics; Natal is the only tropical station for which this requirement is met in most months (Table 2). The variability in ozone is larger in the upper troposphere in the subtropics than in the tropics, and more than 35 soundings are required for 95% confidence intervals of ±15%. For middle latitudes, a minimum of 20 soundings is required for 95% confidence intervals of ±30% near the tropopause, and this will also ensure means reliable to ±15% for 800-500 hPa. These estimates of confidence intervals do not include any bias in the measurements.

The ozone data were analyzed also relative to the altitude of the tropopause. The thermal tropopause was determined from temperature profiles that are provided with each ozone profile; the temperatures are measured by a radiosonde flown with the ozonesonde. The extratropical tropopause may also be defined in terms of potential vorticity [e.g., *Holton et al.,* 1995]. Comparisons of tropopause heights derived from temperatures and from potential verticity may be found in the studies of *Hoerling et al.* [1991] and *Hoinka et al.* [1993]. Thermal tropopause heights were provided for the Canadian stations in the WOUDC archive. For the other stations, the tropopause was determined using the criteria quoted by *Craig* [1965]. At pressures of 500 hPa or lower, the tropopause is defined as the lowest level, with respect to altitude, at which the temperature lapse rate decreases to 2°K km^{-1} or less. The average lapse rate from this level to any point within the next higher 2 km must not exceed 2°K km^{-1}, and the flight must extend at least 2 km above the level. The reliability of determining the tropopause with this procedure is influenced by (1) the vertical resolution with which the data are provided, (2) temperature inversions below the tropopause (particularly for stations from 30° to 40°N), and (3) for high latitude stations, by nearly isothermal profiles that make it unclear where the tropopause is located [e.g., *Bethan et al.,* 1996]. Profiles for which the tropopause determination was clearly wrong, usually due to a temperature inversion, were omitted from further analysis. Fewer than 20 profiles were omitted for most stations, but 10% of the profiles were omitted for Tateno (36°N). For Kagoshima (32°N) the tropopause was required to be at pressures lower than 200 hPa, because of the presence of temperature inversions at lower altitudes. Once the tropopause height was determined, ozone values were interpolated with vertical resolution of 1 km for the region ±6 km from the tropopause, and monthly statistics were formed.

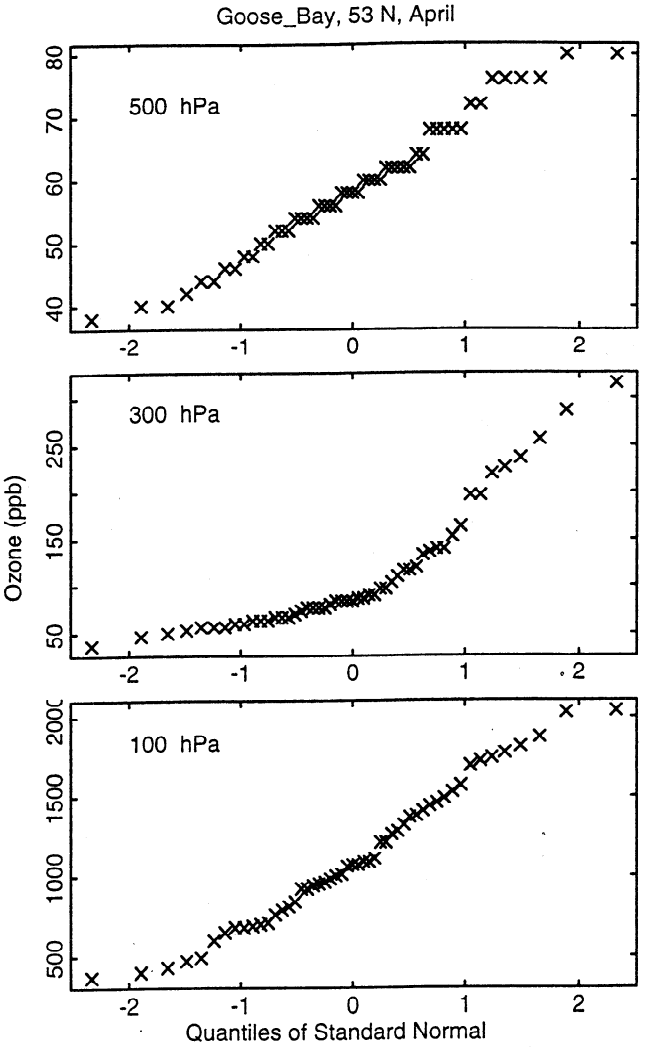


Figure 3. Cumulative probability distribution of ozone values for 500, 300, and 100 hPa for April, at Goose Bay, plotted versus the standard normal distribution. A straight line indicates a normal distribution.

Ozonesondes make measurements near the surface but do not provide the best measure of surface ozone. There is a significant

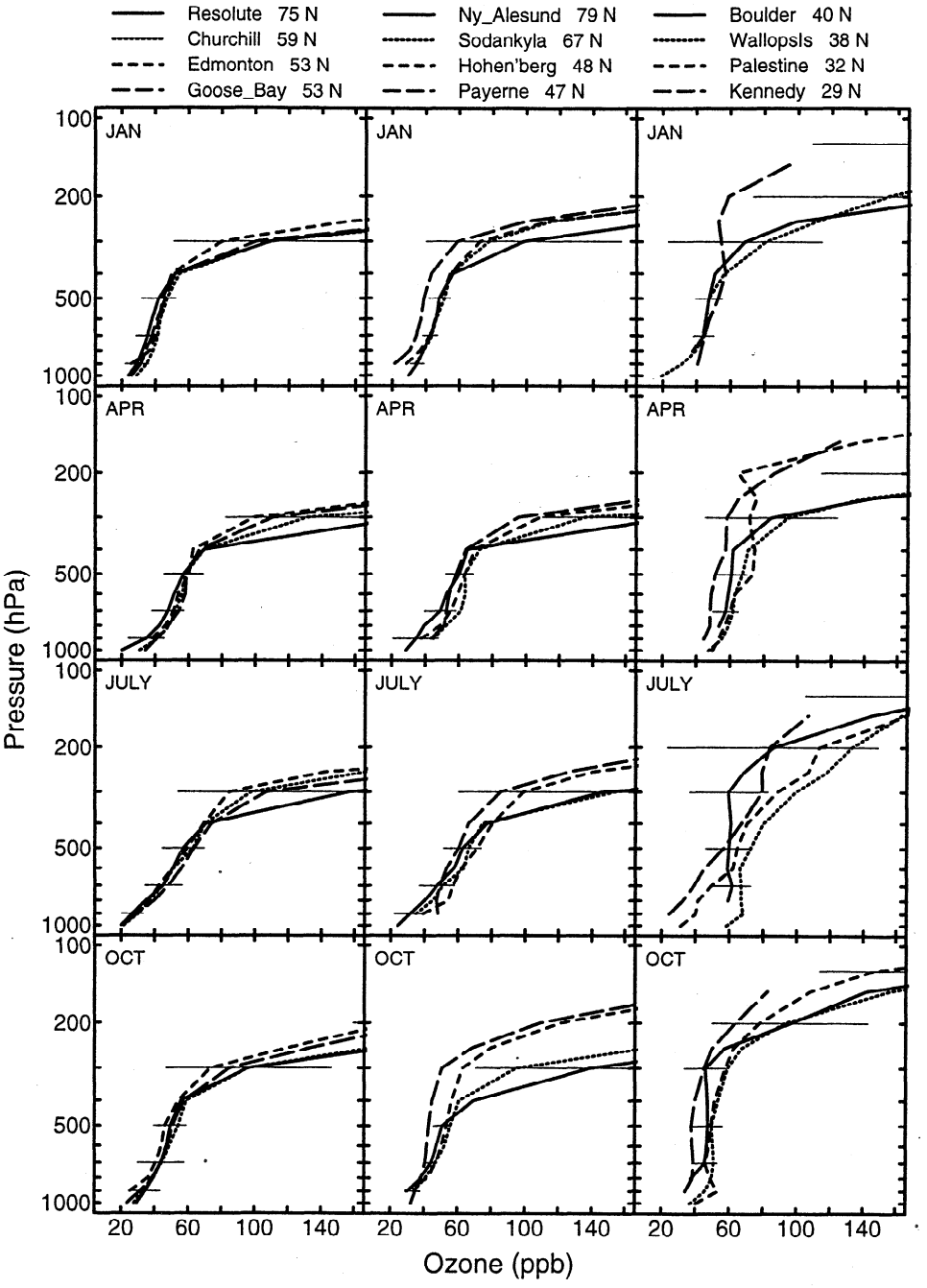


Figure 4. Monthly mean vertical profiles for ozone (in parts per billion) for stations in (left) Canada, (middle) Europe, and (right) the United States. The means are averages of each individual sounding which met the appropriate correction factor criteria for the period given in Table 1. Profiles are shown for January, April, July, and October. The horizontal lines show the mean ±1 standard deviation for selected stations (solid lines) and levels.

diurnal variation in surface ozone at many continental sites, with a minimum in the early morning and a maximum from local noon until late afternoon [e.g., *Logan,* 1989; *Beck and Grennfelt,* 1994]. Many stations release the sondes at 0000 UT or 1200 UT, resulting in a wide range in the local time of measurement. In this study I rely on the analysis of *Oltmans and Levy* [1994] of surface measurements supplemented by my earlier work [*Logan,* 1989] and by data from the literature. I also used data from the WOUDC archive for a few sites: it provides daily means, minima, and maxima.

Ozone residual data were used in combination with the sonde data to evaluate the zonal asymmetry in ozone. I used the tropospheric ozone residual for January 1979 to June 1991, based on version 7 of TOMS data, provided as 3-month mean values for 50°N to 50°S on a grid of 5° in latitude by 10° in longitude [*Fishman and Brackett,* 1997]. Potential errors in the residual are discussed by *Thompson et al.* [1993a], *Hudson et al.* [1995], and *Fishman and Brackett* [1997]. *Fishman and Brackett* [1997] show that the residual generally underestimates the mean tropospheric column measured by the sondes by 15-25%. They attribute this

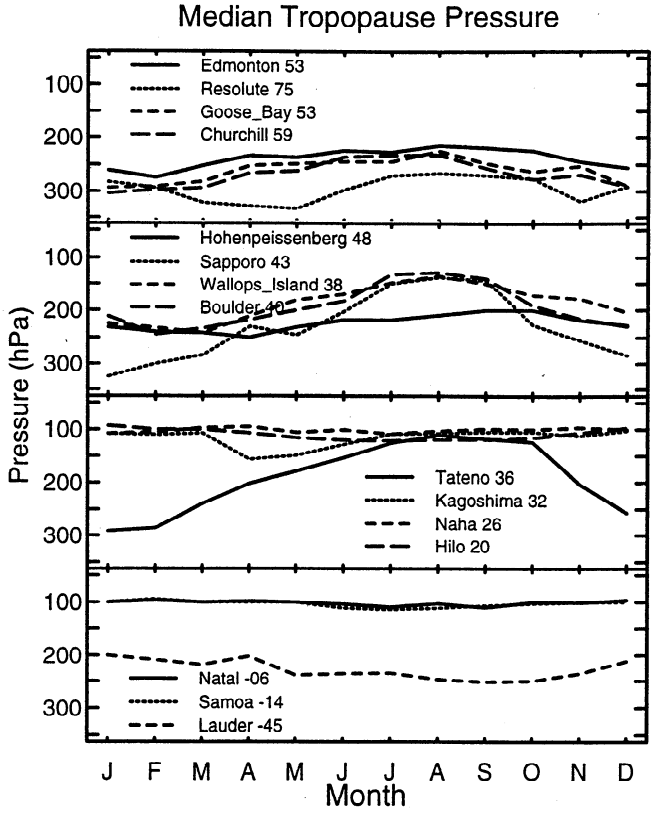


Figure 5. Monthly median values for the tropopause pressures for selected ozonesonde stations.

underestimate to an overestimate by ~2% of the amount of ozone in the stratosphere calculated from SAGE data [cf., *Veiga et al.*, 1995]. I compare the residual with sonde data from the tropics to determine an empirical correction to the residual data, as shown in section 4.

3. Distribution of Tropospheric Ozone

3.1. Northern Middle and High Latitudes

The vertical distribution of ozone is shown for four Canadian stations (53°-75°N) in Figure 4. The mixing ratio increases with altitude gradually from the ground to about 400 hPa (~7 km), then increases much more rapidly around the tropopause. The mean height of the tropopause is between 340 and 220 hPa (8 to 11 km) for these stations (Figure 5). Ozone variability is highest near the tropopause (Figure 6). Monthly mean values of ozone are similar (within 10 ppb) at the four stations except for low values found near the surface at high latitudes in spring, discussed below. Ozone is highest in spring in the lower troposphere (800 hPa), in May to August in the middle troposphere, and in spring in the lower stratosphere (200 hPa), as shown in Figure 7.

Analysis of the data relative to the tropopause height shows that ozone values are highest in summer, June to July, in the upper troposphere and at the tropopause, while they are highest in March to June 2 km above the tropopause (Figure 8). There is no overlap of the central 67% of ozone values at the tropopause with values 1 km above the tropopause except in autumn (Figure 9). The significant change in the phase of the annual cycle of ozone and the steep concentration gradients across the tropopause suggests that high vertical resolution (~1 km) will be required in models to

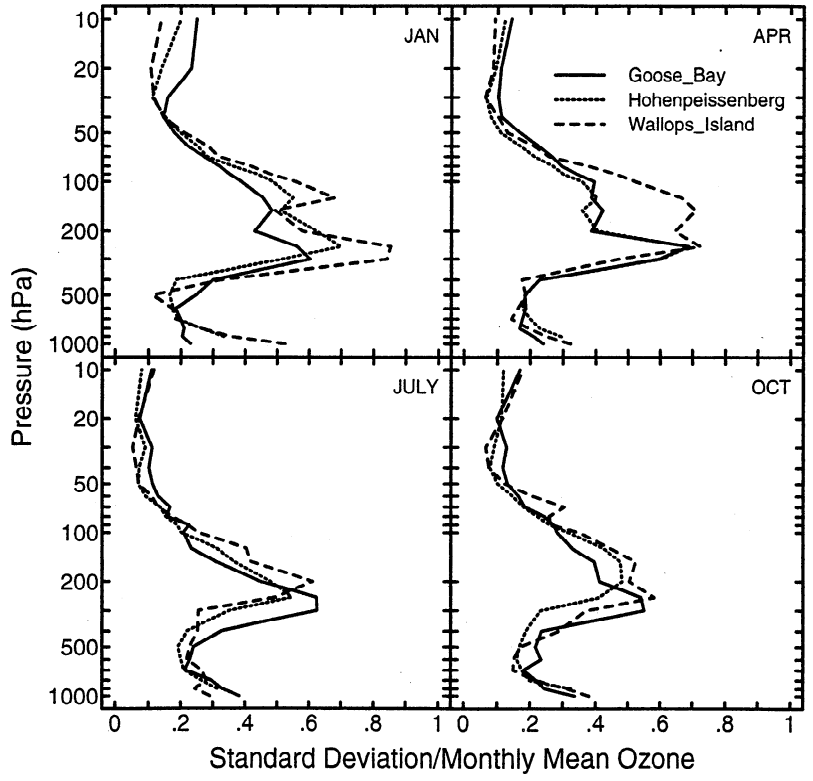


Figure 6. Vertical distribution of the coefficient of variation of ozone for three midlatitude stations for the midseason months.

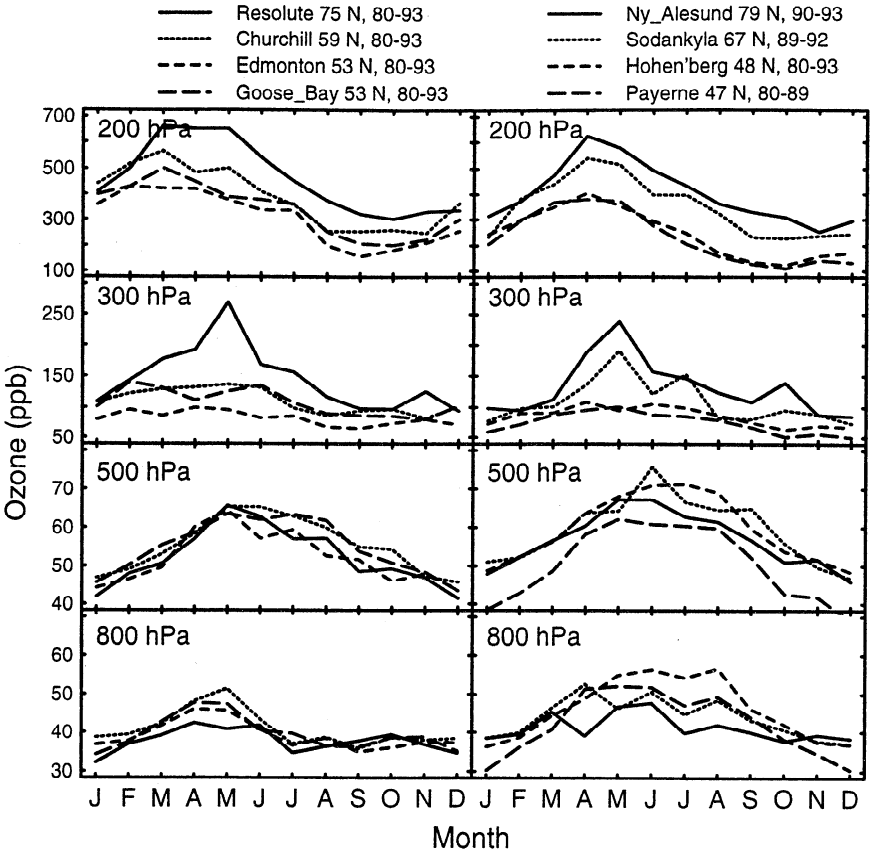


Figure 7. Annual cycle in ozone for 800, 500, 300, and 200 hPa for stations in (left) Canada and (right) Europe.

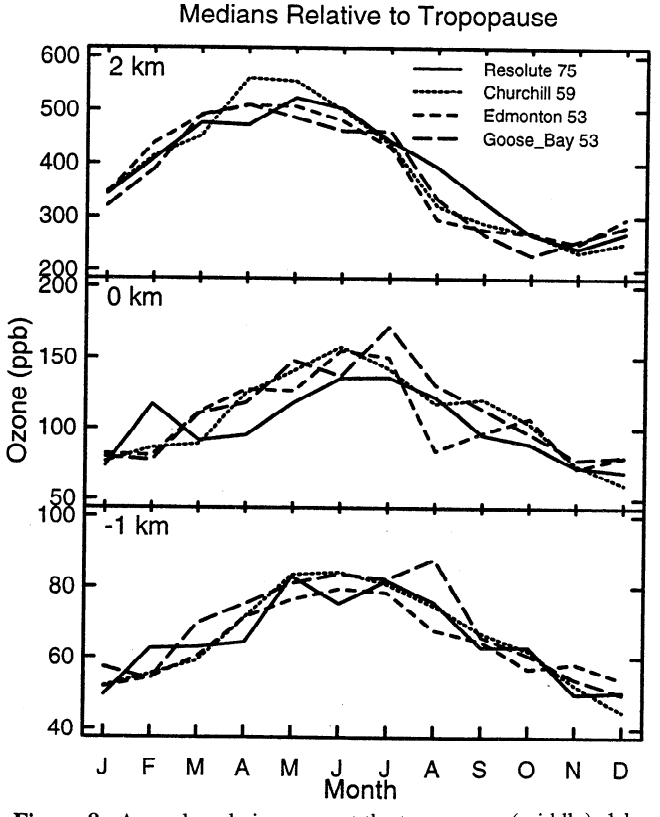


Figure 8. Annual cycle in ozone at the tropopause (middle), 1 km below the tropopause (bottom) and 2 km above the tropopause (top) for four Canadian stations. Monthly median values are shown.

simulate this behavior. Inspection of individual profiles shows that on occasion the ozone partial pressure starts to increase rapidly with altitude before the tropopause is reached [cf. *Bethan et al.*, 1996]. Ozone values at the tropopause vary from tens of parts per billion to a few values over 300 ppb, showing that the thermal tropopause represents a transition between concentrations typically associated with the troposphere (less than ~120 ppb) and those found in the lowermost stratosphere, a few hundred parts per billion. Median values of ozone at the tropopause are 50-100% larger than those 1 km below, while concentrations increase dramatically above the tropopause (Figures 8 and 10). The mean vertical gradient in ozone at the tropopause is steeper by about a factor of 2 when analyzed relative to the tropopause height than when the data are analyzed on pressure levels, because of the large variation in tropopause pressure (Figure 11) coupled with the large vertical gradient in ozone. The standard deviation of ozone at the tropopause is reduced by 15-30% when the data are referenced to the tropopause height rather than to pressure levels, as noted also by *Bojkov and Fioletov* [1997].

Bethan et al. [1996] compared the height of the thermal tropopause with a tropopause defined by the ozone gradient for four sonde stations located from 51°N to 79°N. They defined the ozone tropopause as the lowest altitude where the ozone gradient exceeded 60 ppb/km, with the requirements that the ozone mixing ratio exceed 80 ppb and that the values immediately above the tropopause exceed 110 ppb. They found that the ozone tropopause was ~0.5 km below the thermal tropopause, when the latter is well defined. It is likely that the high ozone values at the thermal tropopause (Figure 9) are those cases where the ozone gradient would imply a lower tropopause than the temperature gradient. The small difference between the two definitions of the tropopause

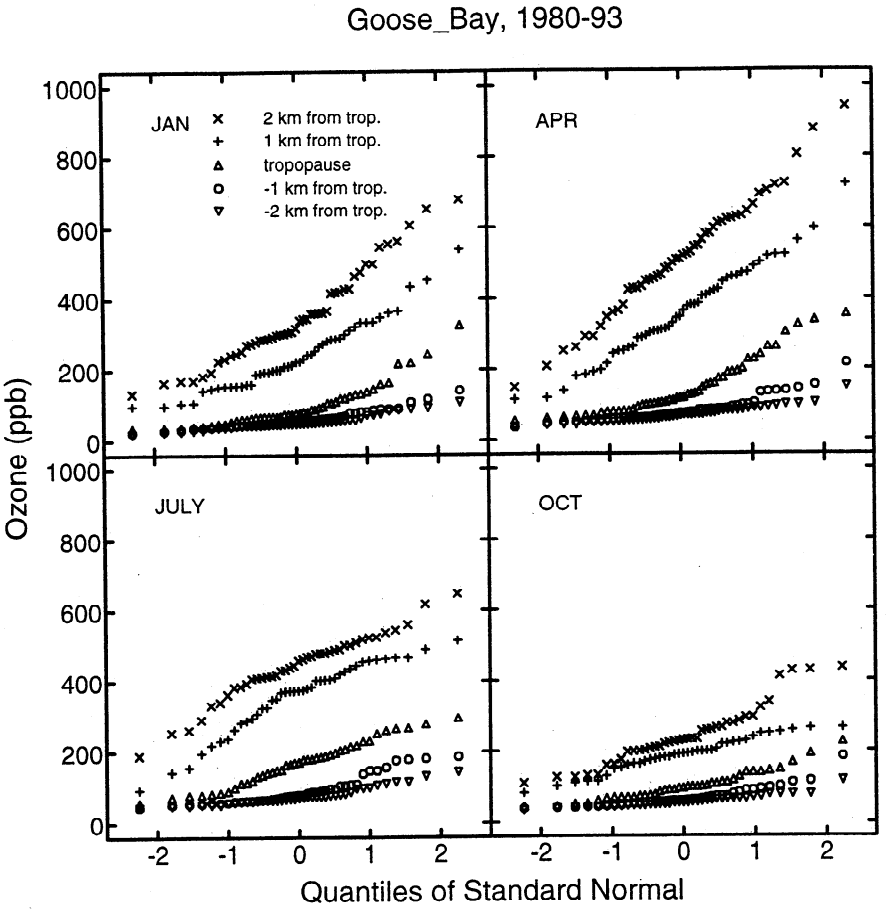


Figure 9. Cumulative probability distribution of the ozone values at the tropopause, ±1 and ±2 km from the tropopause, plotted versus the standard normal distribution. Results are shown for Goose Bay, Canada.

supports the use of the thermal tropopause to provide a separation of tropospheric and stratospheric values of ozone.

Ozone concentrations are highest from April to August from the lower troposphere to the tropopause at the sonde stations in the United States and Europe between ~40° and 70°N (Figures 7, 12, and 13). Median values of ozone at the tropopause are similar over North America and Europe. Ozone values are higher in the lower troposphere over Europe than over Canada by about 10-20 ppb in summer, but the difference is less than 10 ppb at 500 hPa (Figure 7). The differences between the continents near 50°N would be higher if indeed the BM sondes over Hohenpeissenberg underestimate ozone by ~15%, as mentioned above. *Beekmann et al.* [1994b] compared values at five sonde stations in Europe from 44° to 51°N, four BM and one ECC, and found highest values at Hohenpeissenberg and lowest values at Observatoire Haute Provence (44°N); all showed a similar seasonal variation to the results in Figure 7b for 400 hPa and below. Ozone values over the United States near 40°N are higher also than those over Canada in the lower and middle troposphere (Figures 4, 7, and 12). Ozone is generally higher over Wallops Island on the east coast than over Boulder in the middle of the United States; a few percent of this difference is attributed to the use of a different solution strength for KI in the ECC sondes at Wallops Island [*Komhyr et al.*, 1995], while the rest of the difference is likely a true spatial gradient in ozone. Ozone values over Bermuda [*Oltmans et al.*, 1996a] are as high as those over Wallops Island in the middle troposphere in spring and summer (data are lacking for other seasons). Measurements from Palestine show similar concentrations to those over Boulder in the middle troposphere; measurements from Florida in the 1960s show lower values but a similar seasonal cycle, with a summer maximum. There is, however, a summer minimum in the lower troposphere for Palestine, Kennedy, and Bermuda, found also at surface sites in Louisiana and Texas [*Logan*, 1985]. It is attributed to inflow of tropical marine air around the Bermuda High in summer.

Measurements from Japan are shown in Figures 12-14. Similar values are found at all four stations in the winter below 400 hPa, but the seasonal cycle moves from a summer maximum at Sapporo (43°N) to a summer minimum at Naha (26°N), with the minimum becoming more pronounced as the latitude and altitude decrease. The minimum is caused by southwesterly flow of low-ozone air from the tropical Pacific as part of the summer monsoon [*Logan*, 1985]; in winter the prevailing flow is from the Asian continent [*Fukui*, 1977]. The summer minimum in ozone is found also at nonurban surface sites in Japan [*Sunwoo et al.*, 1994]. The high ozone values in the lower troposphere at Tateno are caused by its proximity to Tokyo. The seasonal cycle of ozone over Hong Kong (21° N) is similar to that for Naha [*Chan et al.*, 1998]. Ozone concentrations at the tropopause for Sapporo (43°) and Tateno (36°) are similar to other midlatitude stations in summer but tend to be lower in winter when the tropopause is lower over Japan compared to other midlatitude stations (Figures 5 and 13). The seasonal cycle of ozone at the tropopause is much smaller at the subtropical stations of Kagoshima and Naha, where the tropopause is near 100 hPa.

3.1.1. Surface ozone at northern middle and high lati-

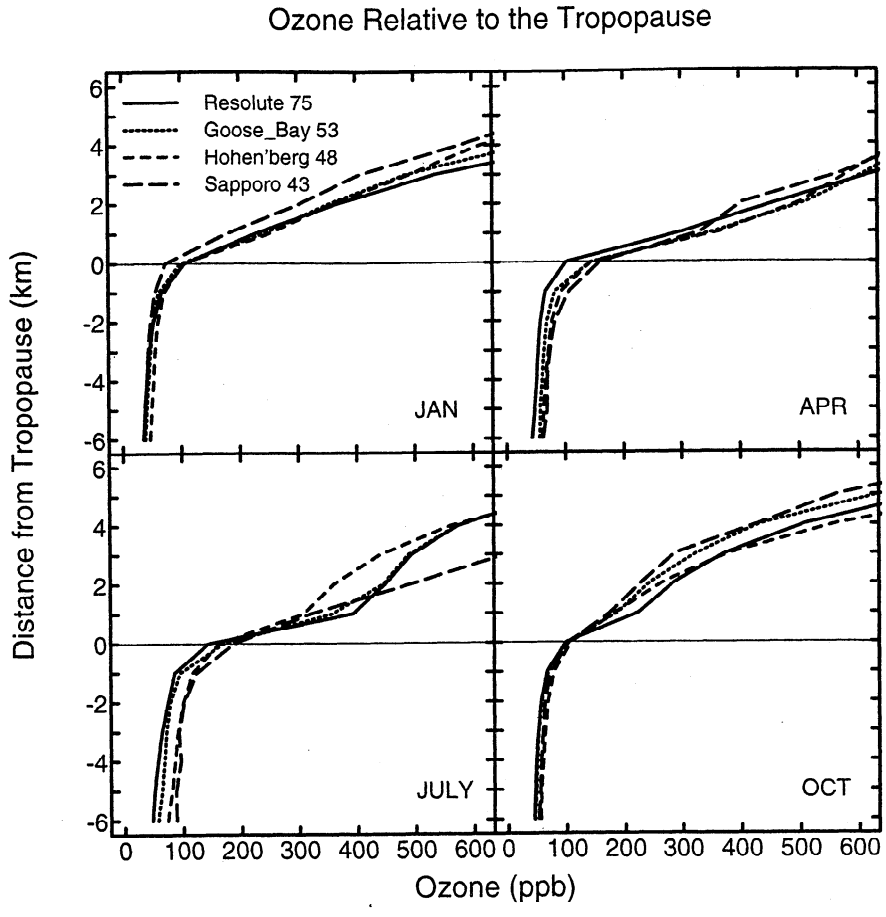


Figure 10. Vertical distribution of ozone relative to the tropopause for four midlatitude stations. Monthly mean values are shown for the midseason months.

tudes. Surface ozone values are highest in spring at remote sites, with the exception of those located in polar regions [*Oltmans and Levy,* 1994], as shown in Figure 15. Surface concentrations are smallest from March to May at Barrow (71° N) and at all the sonde stations north of 75° [*Oltmans,* 1993]. The spring minimum appears to be caused by photochemical loss involving bromine species which occurs over the ice-covered Arctic [e.g., *Barrie et al.,* 1988; *Oltmans et al.,* 1989b; *Fan and Jacob,* 1992; *Mozurkewich,* 1995]. The loss mechanism seems to operate primarily in the boundary layer, and by 850 hPa maximum rather than minimum values are in spring [*Oltmans,* 1993]. There is little diurnal variation in ozone in marine air, and a diurnal variation of only a few parts per billion at remote continental locations [*Oltmans and Levy,* 1994; *Beck and Grennfelt,* 1994]. Ozone concentrations in summer are quite low, about 20-35 ppb (Figure 15) in remote parts of Canada (Bitumount) and northwest Europe (Strathvaich and Mace Head) and the Atlantic Ocean [*Angle and Sandhu,* 1986, 1989; *Winkler,* 1988].

Highest values of nonurban ozone are found in summer at more polluted rural sites in the United States and Europe, and there is a gradient toward higher concentrations from west to east in the United States, and from northwest to southeast in Europe [e.g., *Vukovich and Fishman,* 1986; *Logan,* 1989; *Beck and Grennfelt,* 1994; *Scheel et al.,* 1997; *Fiore et al.,* 1998]. Data from most sites near sea level (except those on the coast) show a strong diurnal variation, particularly in summer. Ozone is depleted in the shallow mixed layer at night by surface uptake on vegetation, and in some locations by local emissions of NO_x. Rural sites downwind of urban areas may have particularly high values of ozone in the middle of the day. Afternoon values of ozone in summer at rural sites are typically 40-70 ppb, while winter values are 20-30 ppb (similar to values at remote sites in winter). This is illustrated in Figure 16 with data from rural sites in the United States, for 1200 to 1600 LT [*Logan,* 1989]; similar results for Europe are given by *Scheel et al.* [1997]; see also Figure 17. Locations with large sources of pollution, but with strong ventilation in summer by cleaner marine air, display a summer minimum in ozone; examples are sites in the southern United States [*Logan,* 1985] and Japan [*Sunwoo et al.,* 1994]. Ozone values in the mid-Atlantic in summer are similar to those at coastal Japanese sites, 22-30 ppb (Figure 17). The various influences on ozone lead to large variations in summer concentrations at the surface at midlatitudes, depending on location, while ozone in the middle troposphere is more uniform.

While the influence of photochemical production of ozone is readily apparent in surface air, it is more difficult to assess the influence of industrial and transportation-related sources of ozone precursors on ozone above the boundary layer. A variety of modeling and empirical approaches have been used. Observations from the Arctic Boundary Layer Expedition (ABLE 3B) allowed quantification of the ozone budget below 6 km in summer. Mean NO concentrations over eastern Canada (in the vicinity of Churchill and Goose Bay) are between 5 and 10 ppt. Model calculations constrained by the observations show that photochemical

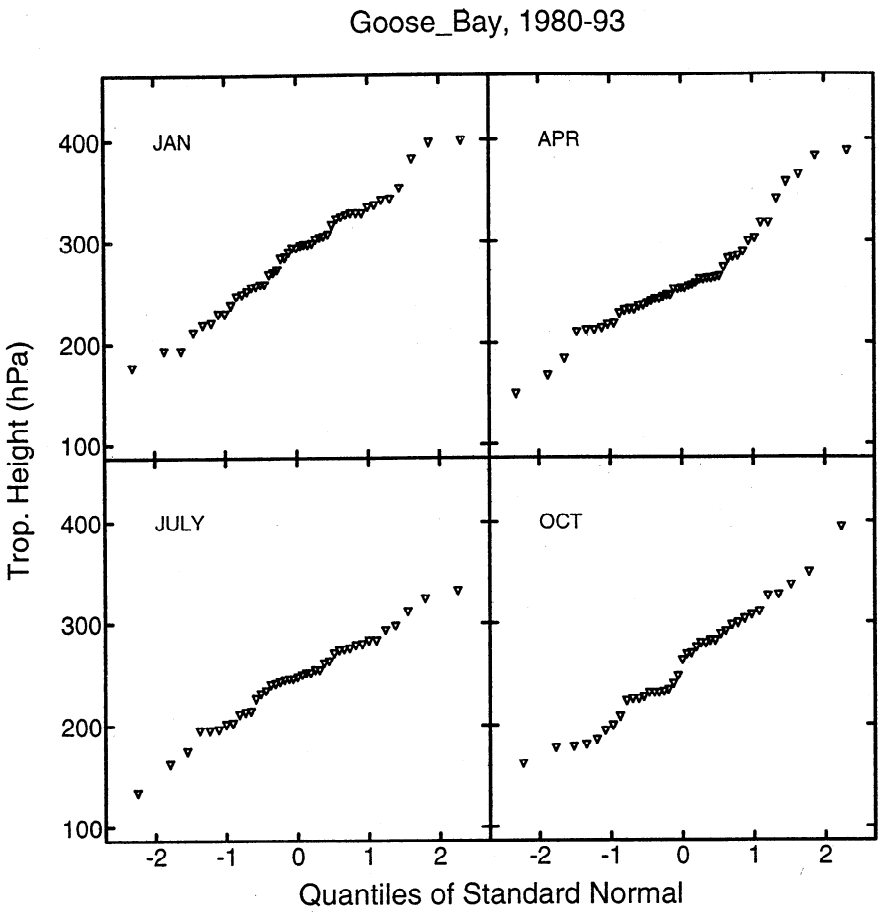


Figure 11. Cumulative probability distribution of the tropopause pressure at Goose Bay, plotted versus the standard normal distribution.

production provides about 60% of the source of ozone below 6 km and that photochemical loss exceeds production by only 15% [*Mauzerall et al.*, 1996]. By contrast, typical NO concentrations at rural sites in the eastern United States and Germany (selected for their relative remoteness) are between 100 and 200 ppt [*Emmons et al.*, 1997]. The polluted continents are clearly net source regions for ozone in summer, as evidenced by spatial gradients in surface ozone discussed above, empirical relations between surface ozone and NO_y or CO [e.g., *Trainer et al.*, 1993; *Parrish et al.*, 1993, 1998] and model calculations [e.g., *Liu et al.*, 1987; *Jacob et al.*, 1993a; *Chin et al.*, 1994; *Liang et al.*, 1998; *Horowitz et al.*, 1998]. Simulations with three-dimensional models indicate that photochemical production is the largest source of ozone at midlatitudes, exceeding the source from the stratosphere by a significant factor in summer [e.g., *Muller and Brasseur*, 1995; *Roelofs and Lelieveld*, 1995; *Wang et al.*, 1998b, c]. *Wang et al.* [1998c] find that the summer maximum in the lower troposphere over the polluted continents is controlled primarily by photochemical production within the continental lower troposphere and that the earlier spring maximum over remote locations is caused by a combination of photochemical production and stratospheric input. They argue that transport of photochemically produced ozone is more effective in spring than in summer because of the longer lifetime of ozone in spring.

3.2. Northern Subtropics and Tropics

Ozone concentrations are highest in spring in the middle troposphere of the tropics and subtropics, in contrast to the behavior at midlatitudes (Figure 18). A spring maximum is evident over Hawaii (Hilo), India (Poona), and the western Pacific (Naha in Figure 12), while a summer maximum is found only over the Bahamas (Grand Turk) in the middle and upper troposphere. The pattern seen over the Bahamas is similar to the behavior found farther north, over Florida and Wallops Island, and is consistent with data for spring and summer months from Bermuda, the Azores, and Tenerife (28° N) [*Oltmans et al.*, 1996a]. Hilo shows a pronounced late summer minimum at 500 hPa and higher altitudes; the minimum extends to the tropopause (Figure 13). Evidently, the seasonal cycle of ozone in the midtroposphere over the subtropics of the north Atlantic is quite different from that over the central and western Pacific. By contrast the seasonal cycle of ozone at the surface is quite similar, with a summer minimum, as shown in Figure 15 by data for Bermuda [*Oltmans and Levy*, 1994] and Japanese islands located near 25°N [*Sunwoo et al.*, 1994].

The spring peak at Hilo in the middle troposphere appears to be caused by a combination of transport of air from Asia leading to photochemical production of ozone and descending air motion from north of 30° which may contribute stratospheric air [*Oltmans et al.*, 1996b; *Wang et al.*, 1998c]. In summer the flow is from the east, and photochemistry probably provides a net sink for ozone [*Harris and Kahl*, 1990; *Oltmans et al.*, 1996b]. Trajectory analysis of selected data from Bermuda and Tenerife shows that in both spring and summer, high ozone values are associated with airflow from higher latitudes and altitudes over North America

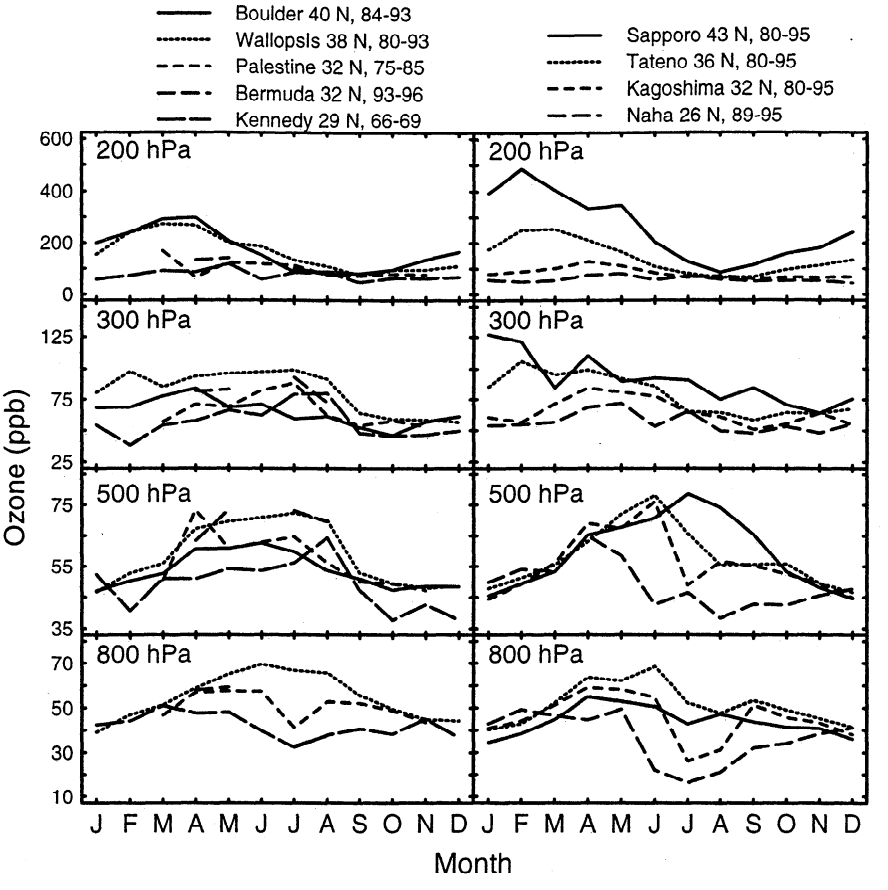
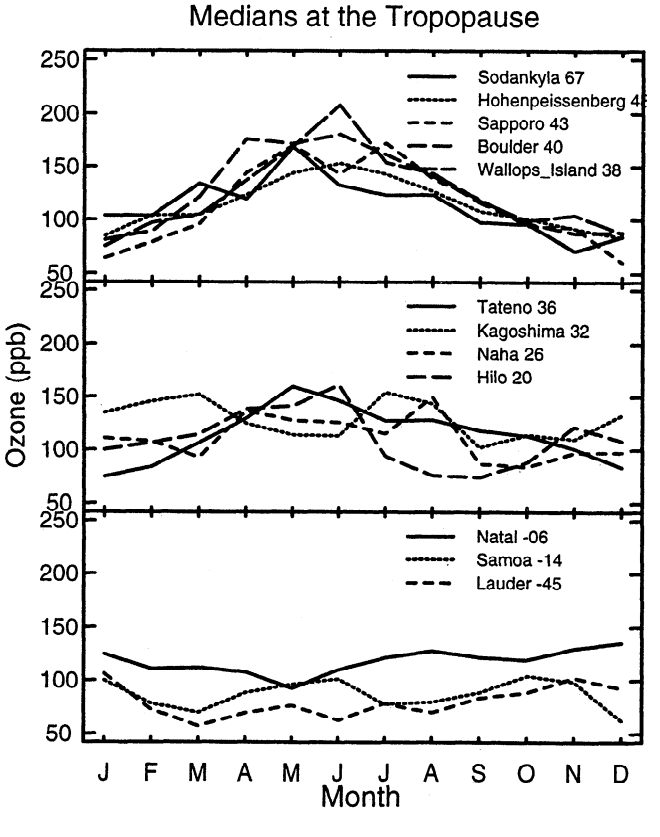


Figure 12. Annual cycle in ozone for 800, 500, 300, and 200 hPa for stations in the United States and western Atlantic (left) and Japan (right). Results are omitted for Boulder at 800 hPa, as the site is at 835 hPa, while the other sites are near sea level. The data from Palestine (32°N) were obtained on an irregular basis in support of balloon campaigns, and measurements are lacking for the winter months.



[*Oltmans et al.*, 1996a]; *Merrill at al.* [1996] suggest that some of the high ozone layers in summer may be caused by stratosphere-troposphere exchange. In the boundary layer the summer minimum is a combination of photochemical loss and tropical airflow [*Oltmans and Levy*, 1992; *Moody et al.*, 1995].

Limited data from Panama (9°N) suggest a spring maximum in the lower troposphere and an autumn maximum in the upper troposphere. Ozone concentrations in this part of the northern tropical troposphere are considerably smaller than those at middle and high latitudes, at all altitudes (Figures 4 and 14). This conclusion is based in part on data obtained in the 1960s, but the general patterns seen in the early data [*Chatfield and Harrison*, 1977a] are borne out by the measurements from Bermuda and Tenerife reported by *Oltmans et al.* [1996a]; a campaign at Panama in July 1977 gave similar ozone concentrations to those measured a decade earlier (Figure 18). Surface concentrations of ozone at Barbados (13°N) [*Oltmans and Levy*, 1994] and Venezuela (9°N) [*Sanhueza et al.*, 1985] are considerably smaller than those at higher latitudes, with a summer-fall minimum and winter maximum, likely reflecting greater photochemical loss in the summer. Measurements made from ships generally indicate low concentrations of ozone (<10 ppb) in equatorial Pacific marine air [e.g., *Piotrowicz et al.*, 1986, 1991; *Johnson et al.*, 1990; *Thompson et*

Figure 13. Annual cycle in ozone at the tropopause. Monthly median values are shown.

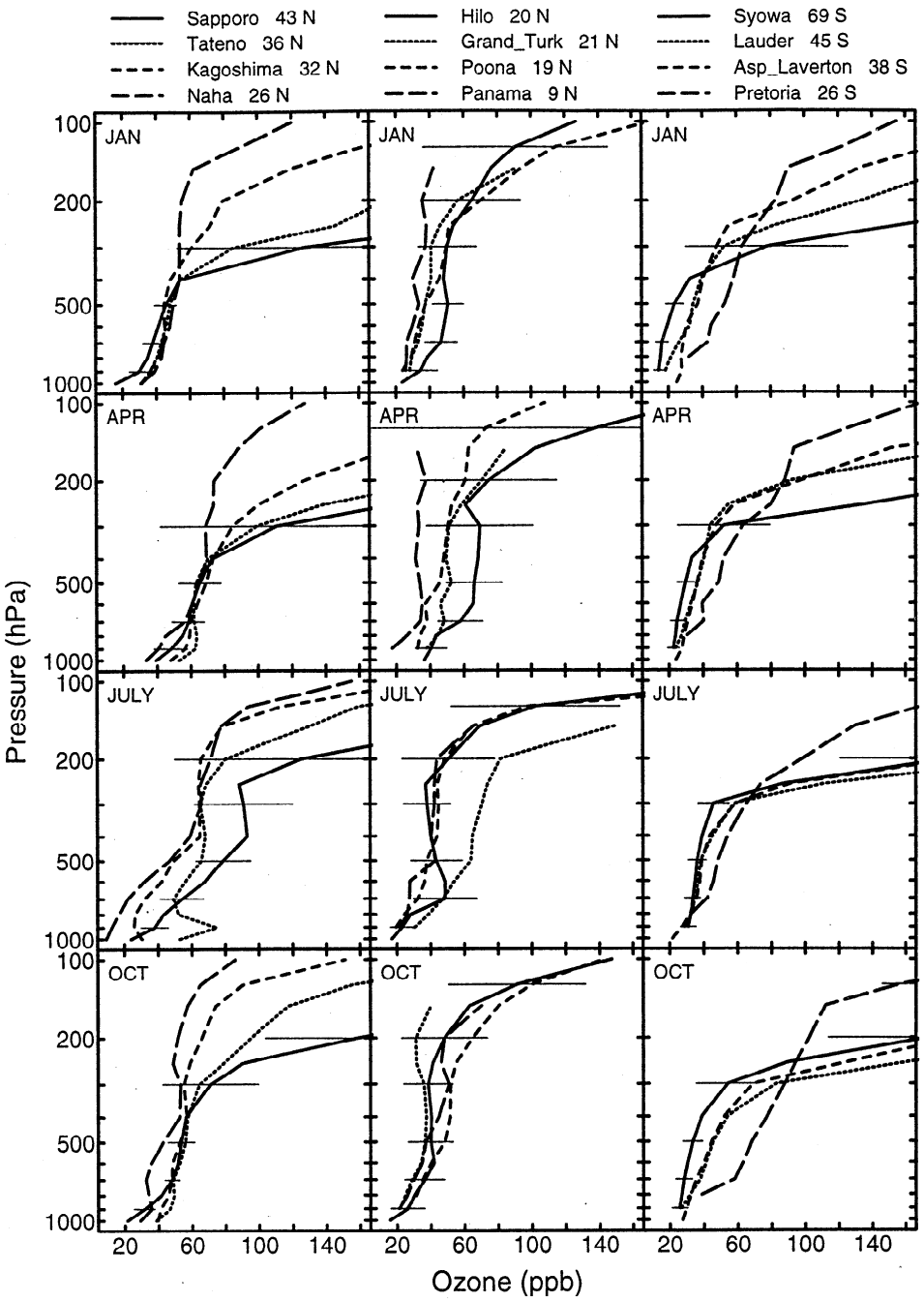


Figure 14. Monthly mean vertical profiles for ozone (in parts per billion) for stations in (left) Japan, (middle) the northern sub-tropics and tropics, and (right) the southern extratropics. See Figure 4 for details.

al., 1993b], as do aircraft data for the tropical marine boundary layer [e.g., *Routhier et al.,* 1980; *Singh et al.,* 1996].

While the data shown here suggest low concentration of ozone in the northern tropics, the measurements are from marine locations. Aircraft data from equatorial west Africa in December, February, and March show ozone concentrations exceeding 70 ppb in the lower troposphere, accompanied by elevated concentrations of CO [*Marenco et al.,* 1990; *Andreae et al.,* 1992]. The high ozone levels were attributed to emissions of precursors by biomass burning during the northern dry season.

3.3. Southern Tropics

The sonde data from the Atlantic and Pacific show the strong longitudinal differences that are apparent in the ozone residual maps of *Fishman and Brackett* [1997]. Lowest values are found year-round at Samoa throughout the troposphere up to the tropopause (Figures 13, 19, and 20). Concentrations at Natal are higher than those at Samoa by less than 15 ppb in May and June but exceed those at Samoa by 10-40 ppb during the rest of the year. Only near the ground do the two stations have similar ozone values for most of the year; Natal receives onshore winds in the lower troposphere [*Logan and Kirchhoff,* 1986]. The measurements at Natal, selected for 1990-1992, are compared to measurements from Ascension Island and Brazzaville for the same years in Figure 20 (right panels). (Ozone concentrations for Natal were somewhat higher for these years than for the longer-term average shown in the left-hand side of Figure 20). Ozone is fairly similar in the middle and upper troposphere for all three stations except

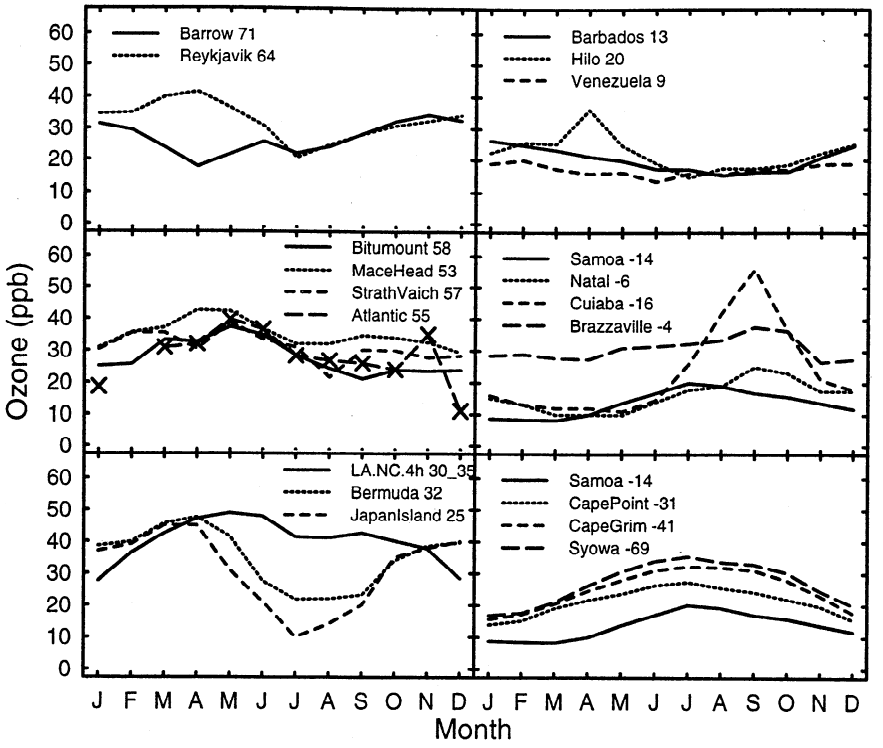


Figure 15. Annual cycle in surface ozone at remote sites. The data are from *Oltmans and Levy* [1994], *Angle and Sandhu* [1988], *Winkler* [1988], *Kirchhoff and Rasmussen* [1990], *Sanhueza et al.* [1985], *Cros et al.* [1988], and WOUDC.

for higher values at Brazzaville in April to June in the upper troposphere. Ozone is highest in June to August in the lower troposphere at Brazzaville but in September to October at Ascension and Natal. Brazzaville has the highest values of ozone in the lower troposphere, with a pronounced maximum in the vertical distribution at 700-800 hPa during June to August. Lowest concentrations of ozone occur in February to May for all stations.

Ozone measurements from Natal, Ascension Island, and Brazzaville are analyzed in detail by *Cros et al.* [1992], *Kirchhoff et al.* [1996], *Nganga et al.* [1996], *Thompson et al.* [1996], *Diab et al.* [1996], and *Olson et al.* [1996], with a particular focus on data obtained during field campaigns in September to October 1992 [*Lindesay et al.*, 1996; *Fishman et al.*, 1996]. These analyses indicate that the high values of ozone in austral spring are caused by

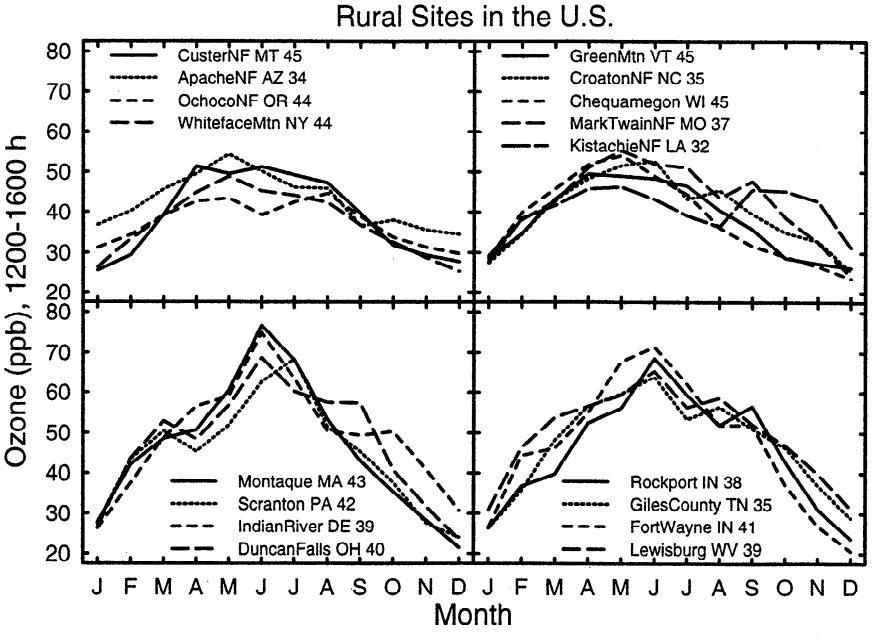


Figure 16. Annual cycle of ozone at rural sites in the United States, for 1200 to 1600 LT. These sites are described in more detail by *Logan* [1988, 1989].

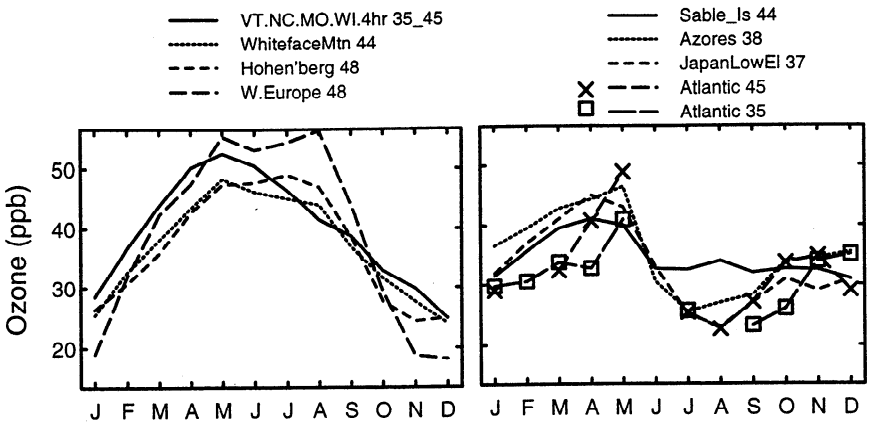


Figure 17. Annual cycle of surface ozone at midlatitudes. (left) The average of four sites in eastern United States (for 1200-1600, Figure 16), Whiteface Mountain (Figure 16), Hohenpeissenberg in Germany (WOUDC data, 24-hour average), and the average of three sites in the center of west Europe (daytime average, *Scheel et al.* [1997, Figure 5e]). (right) Results from Sable Island, in the Western Atlantic, and from the Azores [*Parrish et al.*, 1998], from Japan [*Sunwoo et al.*, 1994], and from the Atlantic Ocean [*Weller et al.*, 1996]; the island data are 24-hour averages.

emissions of precursors from biomass burning in Brazil and southern Africa, combined with favorable meteorological circumstances; near the end of the dry season, convection lifts ozone and its precursors to the middle and upper troposphere, where the air is carried from Brazil over the Atlantic by high-level westerlies, while easterlies in the lower and middle troposphere bring polluted air from Africa [*Thompson et al.*, 1996; *Jenkins et al.*, 1997]. There is slow sinking motion over the Atlantic, where photochemical production exceeds loss in the upper troposphere [*Jacob et al.*, 1996]. The sinking motion gives rise to relatively high values (~ 25 ppb) in the marine boundary layer of the Atlantic in this season [*Heikes et al.*, 1996]. The July maximum in the lower troposphere at Brazzaville (4°S) reflects the peak burning season in May to August in eastern Africa south of the equator, while the secondary peak in January reflects the burning season north of the equator [*Nganga et al.*, 1996]. A secondary peak is evident in January and December also at Natal and Ascension Island, likely reflecting transport of Northern Hemisphere air influenced by biomass burning. The burning season progresses from midwinter to late spring across Africa from east to west, with most fires between July and September and from 5° to 15°S [*Cahoon et al.*, 1992; *Cooke et al.*, 1996; *Justice et al.*, 1996]. The burning season in Brazil occurs primarily between July and October [*Kirchhoff et al.*, 1996], and highest values at Natal occur in August to November.

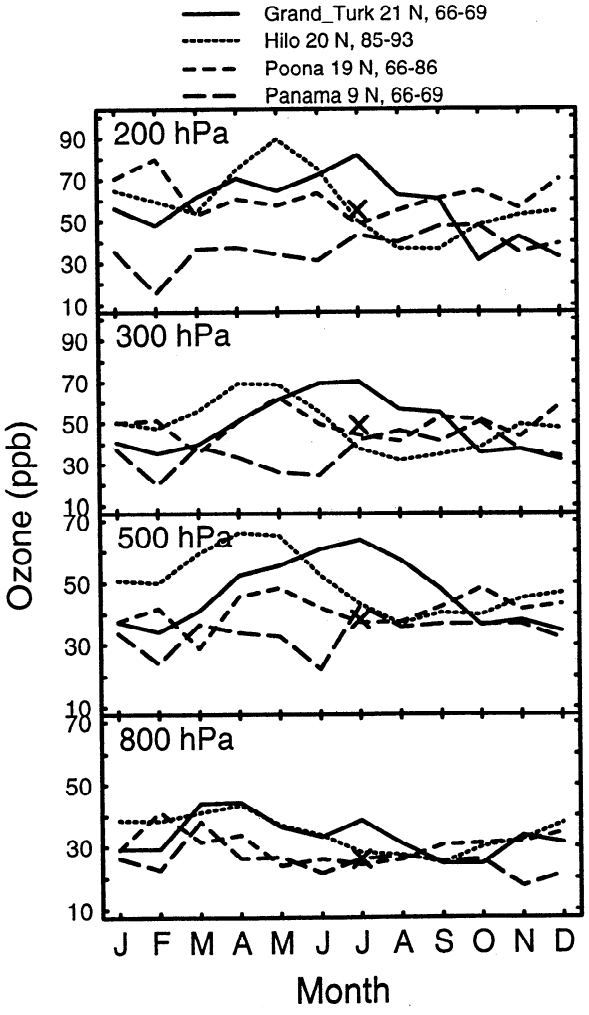


Figure 18. Annual cycle in ozone for 800, 500, 300, and 200 hPa for stations in the northern tropics and subtropics.

The lifetime of ozone with respect to chemical loss is about 3 months in the tropical upper troposphere [*Jacob et al.*, 1996]. The ozone maximum at Samoa in October to November is probably caused by long-range transport of ozone and its precursors from biomass burning, with the peak lagging that at Natal by about a month. Recent analyses of aircraft measurements of trace gases over the southern Pacific confirm the widespread influence of biomass burning in September to October [e.g., *Schultz et al.*, 1998; *Board et al.*, 1999]. Surface ozone at Samoa peaks in July, with a summer minimum, caused by net photochemical loss of ozone in the marine boundary layer; the lifetime for ozone here is only 1 week [*Jacob et al.*, 1996; *Heikes et al.*, 1996].

Limited data from the boundary layer show that ozone can exceed 100 ppb in the vicinity of biomass fires, similar to values found at rural sites in the eastern U.S. during ozone episodes [*Browell et al.*, 1996; *Logan*, 1989]. Mean values at continental locations in Brazil affected by biomass burning are about 50 ppb, as shown in Figure 15 by data from Cuiaba (16°S) [*Kirchhoff and Rasmussen*, 1990; *Kirchhoff and Alvala*, 1996; *Kirchhoff et al.*, 1996]. Typical surface ozone values in the wet season are about 10 to 15 ppb, similar to marine tropical values (Figure 15).

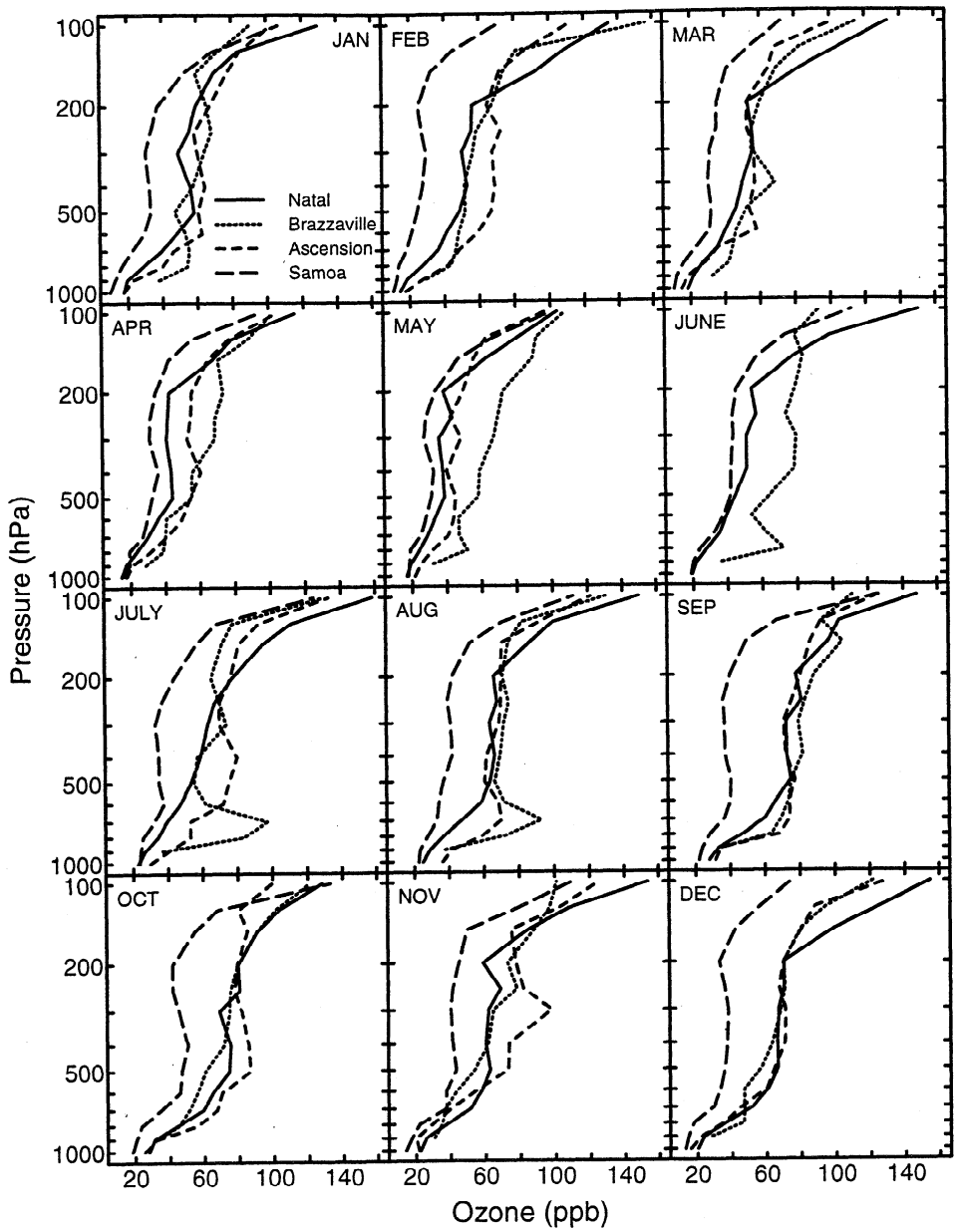


Figure 19. Monthly mean vertical profiles for ozone (in parts per billion) for stations in the southern tropics.

Ozone values in the northern and southern tropics are compared in Figure 21. Concentrations are similar at Samoa (14°S) and Panama (9°N) throughout the troposphere for most of the year, except for the higher values in the lower troposphere at Panama, from January to May. Both stations show higher values in the latter half of the year in the middle and upper troposphere. Ozone is highest in spring for Hilo (20°N) and Natal (6°S); values above Natal are generally higher than those above Hilo in the same season for their respective hemispheres. Ozone values are lowest over Hilo in July to October, and values are similar to those measured over Panama and Samoa in these months.

3.4. Southern Middle and High Latitudes

Observations from extratropics of the Southern Hemisphere are shown in Figures 14 and 22. Ozone is highest in southern spring over South Africa (Irene), Australia (Laverton), and New Zealand (Lauder) from 800 hPa to the upper troposphere but is highest in winter over Antarctica (Syowa). Ozone over Irene is about 20 ppb higher than over Laverton and Lauder in the lower and middle troposphere from August to December. During September and October, Irene is influenced in part by air masses that have recirculated over southern Africa and in part by westerly flow from higher latitudes, as is a site in Namibia where 2 months of data were obtained in 1992 [*Thompson et al.*, 1996; *Diab et al.*, 1996]. This is true also for Reunion (21°S, 55°E) in the Indian Ocean, for which 1 year of sonde data are reported by *Baldy et al.* [1996]. *Fishman et al.* [1991] argue that the influence of effluents from biomass burning on ozone is apparent even at southern midlatitudes, based on the residual data, and the spring maximum at Lauder and Laverton is consistent with this view.

Tropospheric ozone concentrations decrease from 26°S to 67°S in spring and summer but are almost uniform in middle and high latitudes in southern winter, from May to August. Similar uniform behavior is found in winter in the Northern Hemisphere

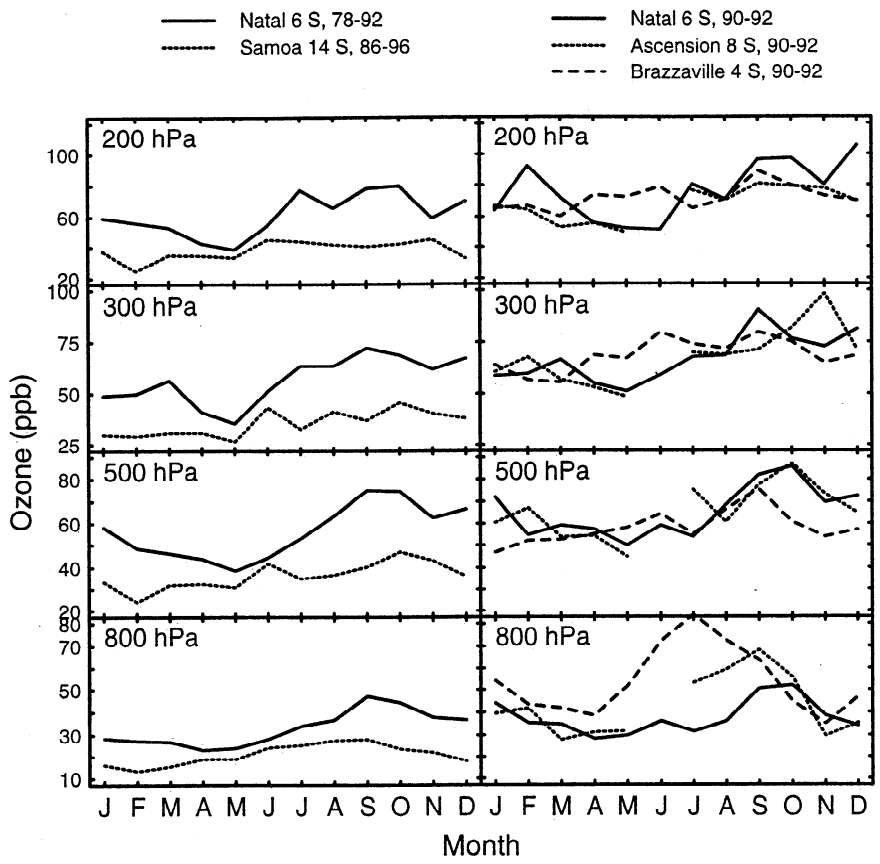


Figure 20. Annual cycle in ozone for 800, 500, 300, and 200 hPa for stations in the southern tropics. (left) Data for Natal for 1978-1992 and Samoa for 1986-1996, (right) data for 1990-1992 for Natal, Ascension and Brazzaville.

(Figures 7 and 12). Ozone values in the upper troposphere are highest in spring at southern midlatitudes but are highest in winter over Antarctica. Ozone concentrations shown for Syowa in Figure 22 for 200 and 300 hPa are significantly lower than pre-ozone hole values, but ozone loss over Antarctica has had little noticeable effect on concentrations below 500 hPa [*Logan,* 1994]. *Oltmans et al.* [1998] find a trend in surface ozone at the South Pole of -0.7% yr^{-1} since 1975.

Observations from remote surface sites are shown in Figure 15. The sites show a winter maximum and summer minimum, with a steady increase in concentration as the latitude increases from Samoa to Syowa. The sites are located in regions expected to have extremely low concentrations of NO_x, and the seasonal cycle is caused in part by larger photochemical loss in summer than in winter [*Ayers et al.,* 1992].

4. Development of a Climatology for Tropospheric Ozone

I present here a climatology for the global distribution of tropospheric ozone derived from ozonesonde data, the tropospheric ozone residual [*Fishman and Brackett,* 1997], and surface observations. The goal is to provide monthly fields of the ozone distribution for use in applications where it is necessary to specify ozone. An earlier version of the climatology was used to calculate the global distribution of OH [*Spivakovsky et al.,* 1990] and in model simulations of the effect of changes in ozone on climate [*Wang et al.,* 1995]. The new climatology is two-dimensional (2-D) at middle and high latitudes, and three-dimensional in the tropics. The sonde data are used to derive zonal averages for the 2-D climatology and vertical distributions for the 3-D climatology, supplemented by surface measurements; the residual data are used to define the spatial distribution in the tropics.

A 2-D climatology for ozone has been developed recently by *Fortuin and Kelder* [1998]. They used ozonesonde data and an earlier version of the tropospheric residual but not surface data.

4.1. Variation of Ozone With Longitude

The zonal asymmetry in the residual is largest between 20°N and 20°S, as shown by *Fishman and Brackett* [1997] and in Figure 23: the sonde data confirm the presence of large gradients in the southern tropics (Figures 19, 20, and 24), with much smaller ozone columns over the Pacific than over the Atlantic and adjoining continents. The residual data are used to derive a 3-D distribution for tropical ozone. The sonde data show little zonal asymmetry for 35°-53°N, except in the lower troposphere in summer (Figures 7 and 12). Values of the residual at the sonde station locations are a few percent higher than the zonal mean value in summer, but there is little bias in other seasons. I elected to use zonal mean values for ozone based solely on the sonde data for 20° to 50°N (with the exception of boundary layer) because (1) the zonal asymmetry is not large; (2) the spatial patterns in the tropospheric column in the residual data do not always match those derived from the sonde data; and (3) the residual column of ozone depends on topography and the local tropopause height. This dependence introduces longitudinal variability in the residual that does not necessarily imply variability in ozone. This is less of an issue in the tropics, where the tropopause height is more constant.

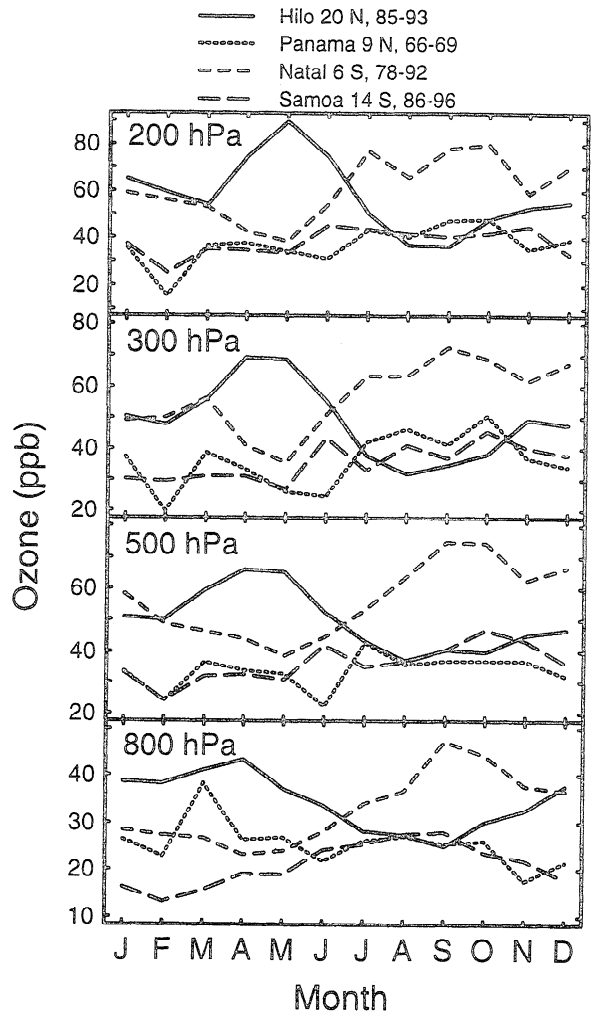


Figure 21. Comparison of the annual cycle of ozone at tropical stations in the Northern and Southern Hemispheres.

Zonal mean values based on the sonde data are used also for 30° to 50°S, again because of the zonal symmetry. The assumption of zonal symmetry is least warranted near 20°N in summer.

4.2. Surface Ozone

Measurements from surface sites were used to specify the concentration of ozone at 1000 hPa. The stations chosen for the climatology are described in Table 3, and the data were shown in Figures 15-17. The data from Barrow were selected for 75°N, while measurements from remote sites in Canada, Iceland, and Ireland were chosen for 60° N and 52.5°N. Ozone values in summer in western Ireland and Scotland are lower than at European continental sites [*Beck and Grennfelt,* 1994] and are similar to values reported for the the mid-Atlantic by *Winkler* [1988].

The majority of year-round surface data for 35°-50°N are from North America, western Europe, and Japan, as discussed above. In summer, ozone values at remote marine locations are much lower than those at rural continental sites, as shown in Figures 16 and 17. These differences are incorporated into the climatology by specifying land and ocean distributions for these latitudes. Surface values over land are mean afternoon (1200-1600) values for four stations on the periphery of the eastern United States, in North Carolina, Vermont, Missouri, and Wisconsin (35°-45°N) [*Logan,* 1985, 1988, 1989]. Summer concentrations for these sites are lower than those for more polluted sites in eastern North America and western Europe but are similar to values at remote sites in the western United States (Figure 16 and 17). No attempt was made to incorporate the spatial distribution of ozone within the United States and northwest Europe [e.g., *Fiore et al.,* 1998; *Liang et al.,* 1998; *Scheel et al.,* 1998], as such data are unavailable for other regions. The observations from marine locations in Figure 17 were used to specify the distribution of ozone over the oceans.

Measurements of ozone over the southern coastal United States are lower in summer than those inland and farther north [*Logan,* 1989; *Fiore et al.,* 1998] and were used to specify land values for 30°N. Data from other continents are lacking for this latitude band. Measurements from Bermuda (32°N) and two Japanese islands near 25° N are similar (Figure 15), with a strong summer minimum in ozone, caused by an influx of tropical air [*Oltmans and Levy,* 1994; *Sunwoo et al.,* 1994]. The data from Bermuda were used to specify marine values of ozone for 30°N. Measurements from lower latitudes (Barbados and Venezuela) show a winter maximum and lower values of ozone. Sonde data from Hilo were taken to represent sea level ozone for Hawaii, as the surface station, Mauna Loa, is at 3.4 km elevation [*Oltmans and Levy,* 1994]. Appropriate averages of these data were used to represent ozone at 30° and 22.5° N (Table 4).

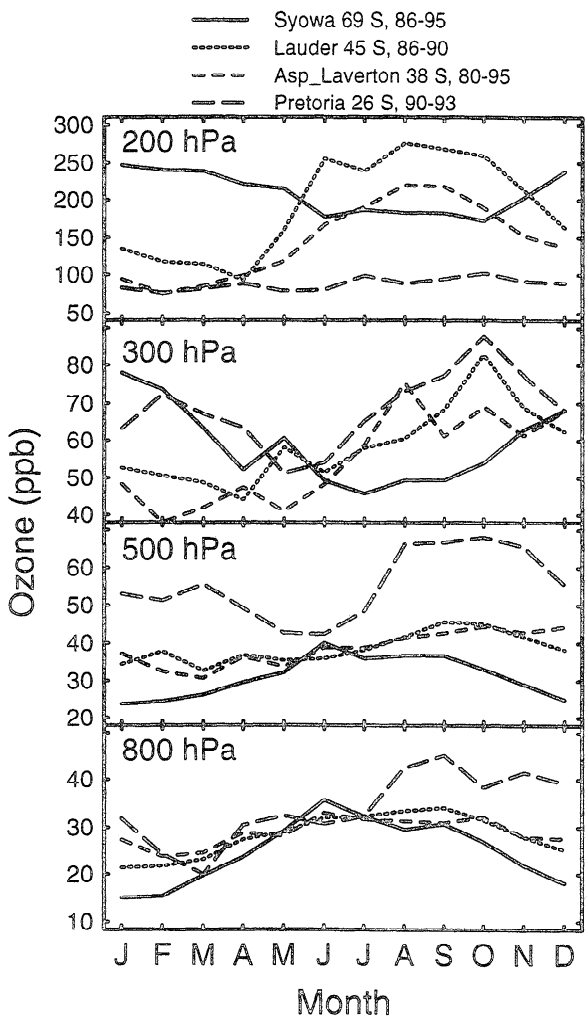


Figure 22. Annual cycle in ozone for 800, 500, 300, and 200 hPa for stations in the southern extratropics.

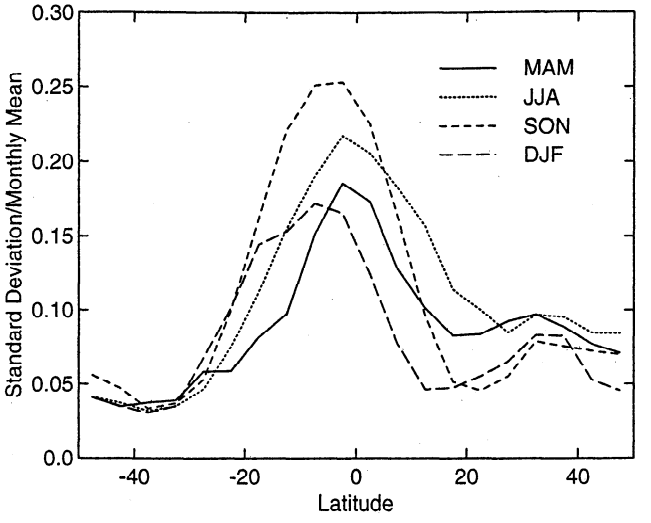


Figure 23. Ratio of the standard deviation to the monthly mean of the tropospheric residual versus latitude. This was calculated from the results given in *Fishman and Brackett* [1997].

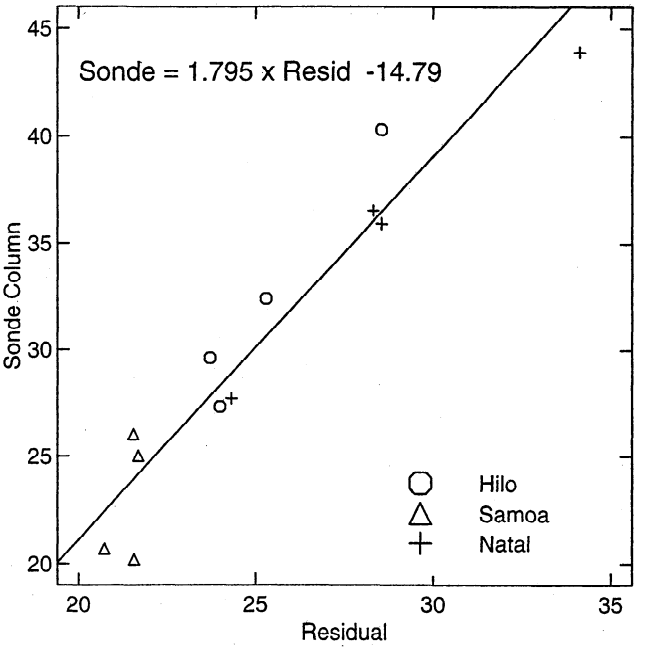


Figure 24. Tropospheric residual (from satellite data) versus the tropospheric column calculated by integrating the ozonesonde data from the surface to the tropopause for tropical sonde stations. The residual was available for 3-month averages. The straight line is a least squares fit to the points, and the text gives the coefficients of the fit.

Ozone profiles in the southern tropics are based on sonde data from Samoa and Natal, so colocated surface data were used to specify ozone for 1000 hPa. Ozone concentrations are low at both these locations and are representative of marine air. Data from continental sites were used for the biomass burning season in the tropics, as discussed below. The limited surface data from southern middle and high latitudes (Figure 15) were taken to represent their respective latitude zones.

4.3. Zonal Mean Climatology

Profiles from sonde stations at similar latitudes were averaged together to produce zonally representative values (Table 4). The results are not true means, given the sparseness of the stations. The latitude resolution was determined by the station locations and by characteristic features of the ozone distribution, including the gradient in the tropopause height with latitude and the behavior of surface ozone. Where sonde data were available from North America, Europe, and Japan, mean profiles were formed for each region, and these were averaged together. The profiles from each station and the resulting zonal means are shown in Figure 25 for January and July.

4.3.1. Northern middle and high latitudes. Ozone profiles are given with resolution of 7.5° for 22.5° to 60° N. Data from three Arctic stations were used to represent ozone at 75°N, while data for Sodankyla (67°N) and Churchill (59°N) were used for 60°N. The European profiles are higher than the Canadian profiles in summer in each case but only by a few parts per billion (Figure

Table 3. Surface Data Used in Ozone Climatology

Station	Latitude	Longitude	Reference
Barrow	71°N	157°W	1
Reykjavik	64°N	22°W	1
Bitumount	58°N	112°W	2
Mace Head	53°N	10°W	1
Sable Island	44°N	60°W	3
VT,WI,MO,NC,4 hours	~40°N	~83°W	4
Azores	39°N	27°w	3
Japanese sites	37°N	~140°E	5
Bermuda	32°N	64°W	1
LA,NC,4 hours	~32N	~85°W	4
Barbados	13°N	59°W	1
Venezuela	9°N	63°W	6
Brazzaville	4°S	15°E	7
Natal	6°S	35°W	8
Samoa	14°S	171°W	1
Cape Point	34°S	18°E	1
Cape Grim	41°S	145°E	1
Syowa	69°S	39°E	1

1, *Oltmans and Levy* [1994]; 2, *Angle and Sandhu* [1986, 1989]; 3, *Parrish et al.* [1998]; 4, Logan [1988, 1989], this paper; 5, *Sunwoo et al.* [1994]; 6, *Sanhueza et al.* [1985]; 7, *Cros et al.* [1988]; 8, *Kirchhoff and Rasmussen* [1990].

Table 4. Data Used in Ozone Climatology

Latitude	Stations
75° N	Mean of Alert, Resolute, Ny Alesund surface: Barrow
60° N	0.33×Sodankyla + 0.67×Churchill. surface: mean of Bitumount, Canada, and Reykjavik, Iceland
52.5° N	0.8×(mean of Edmonton and Goose Bay) + 0.2×Lindenberg(92-95) surface: mean of Mace Head, Ireland, and Bitumount, Canada
45° N	Mean of United States, Europe and Japan, where U.S. is mean of Edmonton, Wallops; Europe is mean of Hohenpeissenberg, Payerne, Biscorosse; Japan is Sapporo surface, land: average based on eastern U.S. rural stations, 1200-1600 LT. surface, ocean: based on Sable Island (except July-September), Atlantic, 45°N 900 hPa (in ppb) is the average of 1000 and 800 hPa (in ppb)
37.5° N	Mean of United States, Europe and Japan, where U.S. is mean of Boulder, Wallops Europe is Cagliari (January-April, September-December); Japan is Tateno Europe is mean of Azores and Cagliari, May-August surface, land, 900, 800 hPa: same values as 45° N, land surface, ocean: average of Azores, Japanese low-elevation sites, Atlantic, 35°N 900 hPa (in ppb) is the average of 1000 and 800 hPa (in ppb)
30° N	Dec.-Feb.: 0.67×Kagoshima + 0.33×Kennedy March, June, Sept.-Nov.: mean of Palestine, Kagoshima April, Aug.: mean of Bermuda, Kagoshima May, July: mean of Bermuda, Palestine, Kagoshima surface, land: average of two U.S. sites, 1200-1600 LT surface, ocean: Bermuda 900 hPa (in ppb) is the average of 1000 and 800 hPa (in ppb)
22.5° N	Mean of Naha, Hilo, and Grand Turk surface: mean of Bermuda (32° N), Barbados (13° N), and Hilo (1000 hPa)
12° N - 20° S	For vertical profiles, use Samoa or Natal, depending on values of residual DJF, MAM: Samoa for <26 DU, Natal for >26 DU JJA: Samoa for <30 DU, Natal for 30-40 DU, mean of Ascension, Brazzaville for >40 DU SON: Samoa for <30 DU, Natal for >30 DU surface: Samoa and Natal surface measurements, supplemented as in text
26° S	0.85 × Irene values for 700-200 hPa surface: Cape Point
37.5° S	Aspendale/Laverton surface: Cape Grim
45° S	Lauder surface: Cape Grim
67° S	Syowa

25). I used recent data taken with ECC sondes at Lindenberg to represent Europe at 52.5°N; concentrations are 10-20 ppb higher than those at the Canadian stations in April to October. Since the majority of land at 52.5°N is in remote areas, the Lindenberg data were weighted by only 0.2 in forming the zonal average.

There is a significant gradient in tropospheric ozone over North America from 53°N to ~40°N, as evident in Figures 4, 7, and 12. Data from Edmonton and Wallops Island were used to form a North American average for 45°N. In the lower troposphere these stations represent a remote location with low ozone, and a site affected by regional air pollution, particularly in summer, with higher ozone (Figure 25d). Data from Boulder were not used for 45°N, because of the higher tropopause in summer, and the effects of pollution from Denver at 800 and 700 hPa. Results from three stations were used for Europe. Data from Biscarosse are for 1976-1983, but measurements from nearby Observatoire Haute Provence for 1984-1990 [*Beekmann et al.*, 1994b] show similar values. The results from Sapporo, Japan, were weighted equally with the mean profiles for North America and Europe to form a mean for 45°N. A similar procedure was used for 37°N (see Table 4), but there were measurements from only one location in Europe, Cagliari, for 1968-1980; profiles for Cagliari are similar to those for Biscarosse and Payerne. Recent data from the Azores were combined with the Cagliari data for May to August. Many of the sonde stations from 38°N to 48°N are influenced by regional air pollution. The surface data for 1000 hPa (see above) and for 800 hPa at 45°N were averaged to give values at 900 hPA. The concentrations for 1000, 900, and 800 hPa at 45°N were used also at 37.5°N, as the data for Tateno, Boulder, and Wallops below 800 hPa are unlikely to be typical of the latitude zone, since all are affected by regional pollution.

4.3.2. Northern subtropics and tropics. The sparseness of data makes it more difficult to define representative profiles for lower latitudes (Figure 1), and the profiles used in the climatology are more uncertain than those for higher latitudes. Kagoshima is the only station near 30°N with more than 15 soundings in most months (Table 2). The profiles for 30°N are based on data from Kagoshima, Palestine, and Bermuda supplemented by 1960s data from Florida for the winter. Data are available for Bermuda for 4 months, while Palestine is lacking winter data and has few profiles in several months. Consequently, different stations were used for different months as shown in Table 4. The three stations used to represent 22.5°N are in regions near the maximum in the residual (Grand Turk, 1960s data) and near the minimum (Hilo and Naha) for this latitude in summer, and there is a wide range of ozone values at these stations (Figures 14 and 18). A simple average was adopted for 22.5°N.

4.3.3. Southern subtropics and midlatitudes. The station in

South Africa (Irene) provides the only 3-year data set for the vertical distribution of ozone in the subtropics. The residual in the Irene grid box is ~6% larger than the zonal mean. Irene is located 1.5 km above sea level, which biases the residual low compared to most of its latitude band. Taking these factors into account, the mean profile for 26° was formed by reducing the ozone values for Irene by 15% for 700-200 hPa and using the Cape Point data for 1000 hPa. The climatology for higher latitudes in the Southern Hemisphere was based on the single station available for each (Table 4).

4.4. Three-Dimensional Climatology in the Tropics

The residual data were used to derive a 3-D distribution from 12°N to 20°S. Comparison of the residual with the integrated column of ozone for three tropical sonde stations with year-round data indicates that the residual overestimates tropospheric ozone and that there is a linear relationship between the two measures of the tropospheric column (Figure 24). The regression line between the sonde and residual columns was used to scale all tropical values of the residual above 20 Dobson units (DU).

The vertical profiles of ozone were determined from monthly data for Samoa (14°S) and Natal (6°S). These stations were selected because they have more year-round data than other tropical stations. There are no tropical stations in the Northern Hemisphere with adequate data to define monthly profiles (Table 2). Profiles from Samoa were used for lower values of the residual (less than 26 or 30 DU depending on season, Table 4), and profiles from Natal for higher values, with the boundary chosen such that the Samoa profiles were used for most of the Pacific Ocean. The shapes of the ozone profile for Brazzaville and Ascension Island are substantially different from those for Natal in June, July, and August (Figure 19), so the mean profile for these two locations was used for ozone residuals greater than 40 DU in these months. Gridded values of ozone were calculated by dividing the monthly sonde profile by the seasonal column derived by integrating the sonde data and multiplying it by the residual value for that grid box.

The 3-D climatology was derived with resolution of 4° in latitude by 5° in longitude. The procedure described here was used for 12°N to 20°S. Zonal means (Table 4) were interpolated every 4° for 20° to 90°N and for 24° to 90°S, allowing for different values at 1000 and 900 hPa for land and ocean for northern midlatitudes as discussed above. Intermediate latitudes (12°-20°N and 20°-24°S) were determined by interpolating between the 2-D and the 3-D distributions to give a 3-D distribution.

Surface data from Brazil and Africa indicate that the low ozone values measured at Samoa and Natal (typical of marine air) are much lower than continental values measured during the biomass burning season, as illustrated by data from Cuiaba (Figure 15). Surface and sonde data from sites in Brazil [*Kirchhoff and Rasmussen,* 1990; *Kirchhoff et al.,* 1996] and surface data from southern Africa [*Cros et al.,* 1988; *Thompson et al.,* 1996] were used to derive typical values of ozone to use in the lower troposphere in regions affected by biomass burning; aircraft data were used to derive values for northern Africa [*Marenco et al.,* 1990; *Andreae et al.,* 1992]. These values are given in Table 5. Maps of fire counts [*Arino et al.,* 1997] were then used to determine regions where these values should be used in the climatology. Increasing the ozone values in the lowest layers to take into account the contribution of biomass burning changes the tropospheric ozone column by only a few percent. Continental values of ozone in the tropics are similar to marine values during months without biomass burning, as shown by data for Cuiaba (Figure 15) and Manaus [*Kirchhoff et al.,* 1988, 1990].

4.5. Summary of the Climatology

The vertical distribution of ozone derived for the zonal climatology is shown in Figures 25 and 26 for 22.5°N to 75°N, and the annual cycle is given in Figure 27. The spatial distribution of ozone is shown in Figures 28a and 28b and the distribution with latitude is shown for the mid-Pacific (170°W) and mid-Atlantic (30°W) in Figures 29a and 29b. There is almost no gradient in zonal mean ozone values from 30° to 75° in the winter hemispheres for 800 to 400 hPa, while in summer the spread of mean ozone values is 10 ppb, with highest values for 37°-50°N. Winter values of ozone at northern midlatitudes are 5-10 ppb higher than those at southern midlatitudes in the middle troposphere, while summer values are 20-30 ppb higher in the north than those in the south. Highest values of ozone at 500 hPa (~6 km) are found at northern midlatitudes in April to July, 65-70 ppb, while they are found over the south tropical Atlantic for August to January, 65-85 ppb. Lowest values, 25-30 ppb, are found over the equatorial Pacific year-round and at southern high latitudes in November to March.

Table 5. Ozone Values (ppb) Used in Regions of Biomass Burning

	South America					
hPa	July	Aug.	Sept.	Oct.		
1000-800	26	36	45	36		
	South Africa					
hPa	July	Aug.	Sept.	Oct.	Nov.	Dec.
1000-800	31	36	45	38	35	26
	North Africa					
hPa	Nov.	Dec.	Jan.	Feb.	Mar.	
1000	20	20	20	20	20	
900	40	50	50	40	40	
800	55	70	70	55	40	
700	40	70	70	40	40	

These values are used to overwrite the ozone values in specified geographic regions of the ozone climatology, if the ozone value is lower than the values given above.

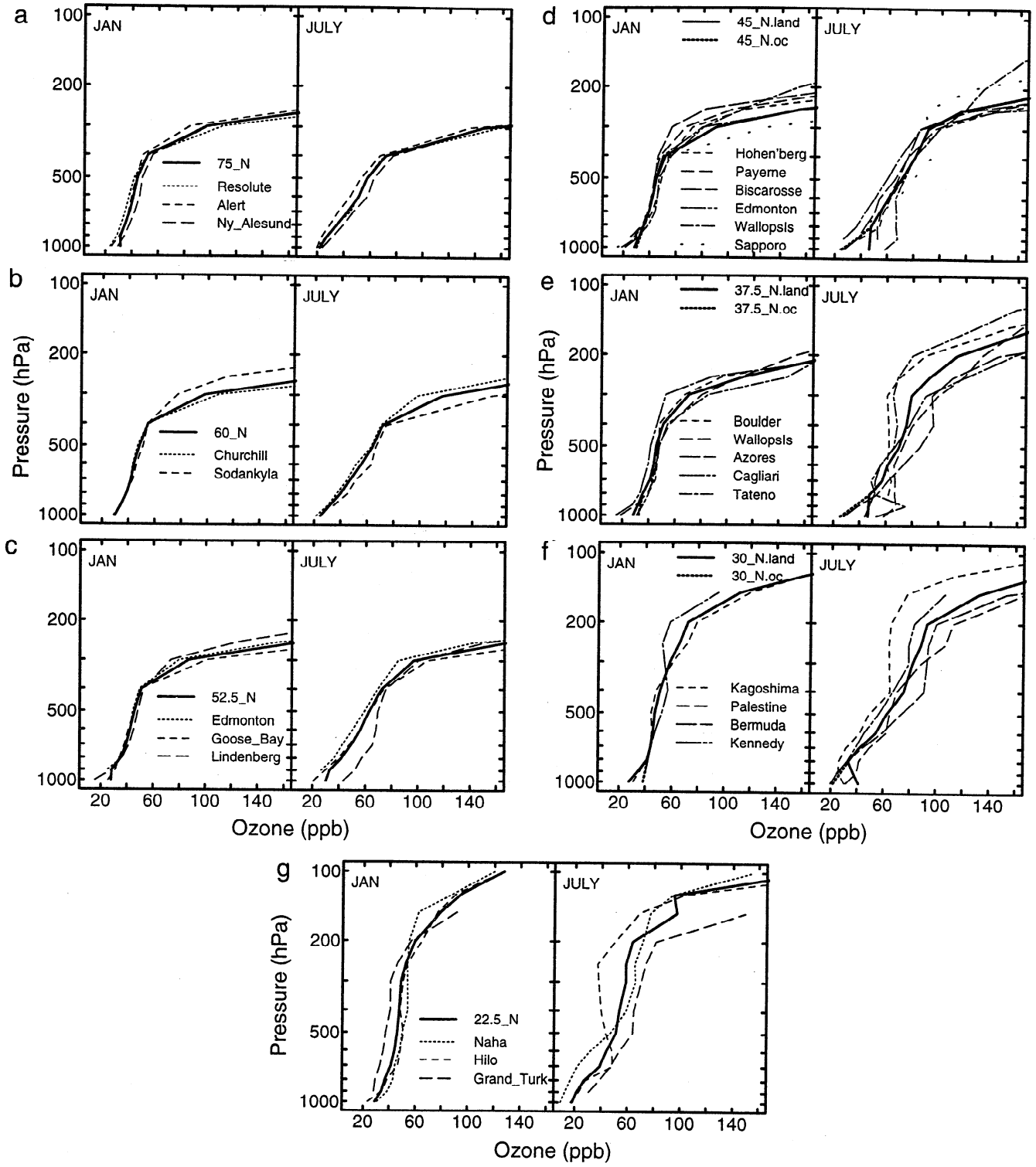


Figure 25. (a-c and g) Thick solid lines show the ozone profiles derived for the zonal mean climatology for January and July for 52.5° to 75°N and for 22.5°N; (d-f) dashed thick lines show values used for the oceans for 1000 and 900 hPa, and the thick solid lines the values used over land for 30° to 45°N. The thin lines show the mean values for the stations used in forming the zonal mean as described in Table 4.

4.5.1. Errors in the climatology. At least four kinds of errors or assumptions contribute to uncertainties in the climatology: (1) measurement errors, (2) variability in ozone at a given location, which may or may not be captured by the available data, (3) the assumption of zonal symmetry for ozone for 20° to 90° (station-to-station variability), and (4) the assumption that the station locations are representative of their regions. As discussed in section 2, most stations use ECC sondes for which the bias is ±5%. One standard error is typically 4% (3-7.5%) of the monthly mean ozone value for 800 to 400 hPa at middle and high latitudes (Figure 2). The station means that were used to form zonal means for 20°-75°N are within 15-20% of the zonal mean in summer and

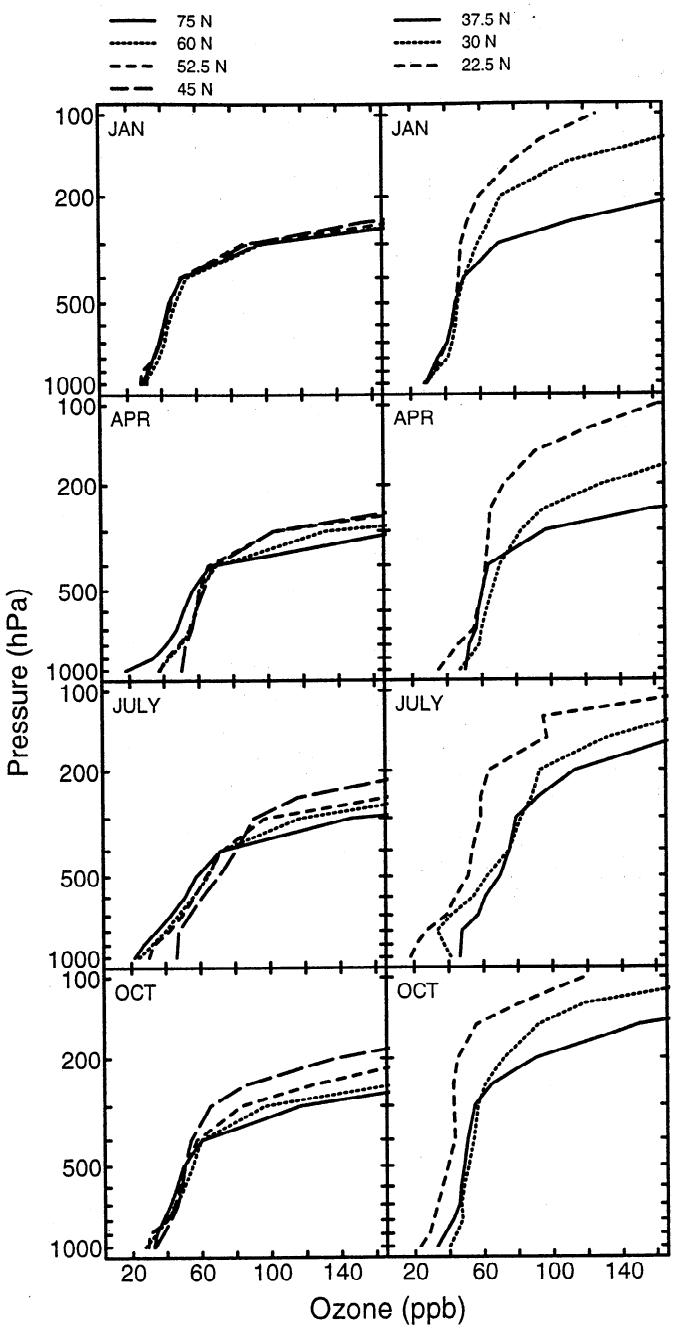


Figure 26. Vertical profiles of ozone for 22.5° to 75°N in the climatology.

within 10% in winter for high latitudes, within 20-30% in summer and 10-20% in winter for middle latitudes, and within 20-35% in summer and 25% in winter in the subtropics (Figure 25). These are maximum differences from the zonal means (not standard deviations). Combining a measurement bias of ±5%, 95% confidence intervals for an individual station monthly mean of ±8%, and a between-station error of 25% leads to a overall error (root-mean- square) of 27% for northern midlatitudes in summer. The error is dominated by the zonal asymmetry. Errors should be smaller in winter when the zonal asymmetry is less and should be larger in the subtropics in summer when the asymmetry is more; also, the standard errors for individual stations are larger in the subtropics (Figure 2). Errors are also smaller at high latitudes. There are errors in surface ozone values, particularly at northern midlatitudes; the division into continental and marine values allows for some of the spatial variability in boundary layer ozone. Continental ozone is too low over polluted portions of North America and Europe by as much as 20-50% in summer but may be too high over remote regions of Asia, where data are lacking.

It is more difficult to quantify errors for the tropics, because the distribution is based on the tropospheric residual combined with vertical profiles, the shapes of which are assumed to be typical. There are likely to be regions of the tropics for which these shapes may not be appropriate, for example in regions of biomass burning, although this is compensated for in those parts of South America and Africa for which additional information is available. One standard error is 5-15% of the monthly mean ozone value for Samoa and Natal, where there are fewer measurements than for extratropics (Table 2). There are errors in the residual itself. The TOMS data likely underestimate the amount of ozone in the lower troposphere over cloud free areas [e.g., *Hudson et al.,* 1995], and the comparisons of residual data and sonde data indicate an underestimate for most stations [*Fishman and Brackett,* 1997; see also Figure 24]. The residual values were adjusted upward for the climatology based on the comparison with sonde data shown in Figure 24. Using the fit in Figure 24, the adjusted residual is within 18% of the seasonal means for Samoa and within 10% for Hilo and Natal. Clearly, errors could be larger, but data are lacking to evaluate them further. Combining errors of ±5% in sonde bias, 20% for 95% confidence intervals for the profiles, and 20% for the residual leads to a overall error of ~30%.

The climatology is likely to be most uncertain in the northern tropics, since no in situ data were used for its derivation. The vertical distribution of ozone was measured on four Atlantic cruises from 50°N to 50°S [*Smit et al.,* 1989; *Weller et al.,* 1996], and these were compared with the climatology. In the northern tropics the climatology is somewhat lower than values measured in January/February (by up to 10 ppb), in good agreement for March/April and for 10°-20°N for May/June, but too low by up to 20 ppb for 10°S-10°N, and it is too high by up to 10 ppb in October/November. The profiles obtained in the Atlantic represent a snapshot in time and are always within 25-30% of the climatology, if not better. The climatology is also in good agreement with ozone values in the marine boundary layer measured on aircraft over the tropical Atlantic [*Heikes et al.,* 1996] and Pacific [*Schultz et al.,* 1998].

In southern subtropics and midlatitudes the climatology is based on data from single stations and the assumption of zonal symmetry. The residual data suggest that the latter assumption is reasonable for 30°-50°S, with relative standard deviations of 4-5% (Figure 23). Preliminary sonde data from Easter Island (27°S) indicate that values are generally smaller than those at Irene (except in May and June) by 5-20 ppb, 10-35%. Values for the climatology are those from Irene reduced by 15%. One standard error of the monthly means for the stations from 38° to 70°S is 4-8%. Combining errors of 5% for bias, 12% for 95% confidence intervals for the profiles, and 5% for zonal asymmetry leads to an overall error of 14%. Errors are likely to be larger in the subtropics where it was necessary to rely on limited data from Irene.

5. Evaluation of Models With Ozonesonde Data

The sonde data presented here are sufficient to describe the abundance and annual cycle of ozone reasonably well in many regions of the world, with the northern tropics and the equatorial Pacific being obvious exceptions. Measurements from the majority of stations given in Table 1 are suitable for testing 3-D models of ozone. Monthly statistics for each station are provided for a

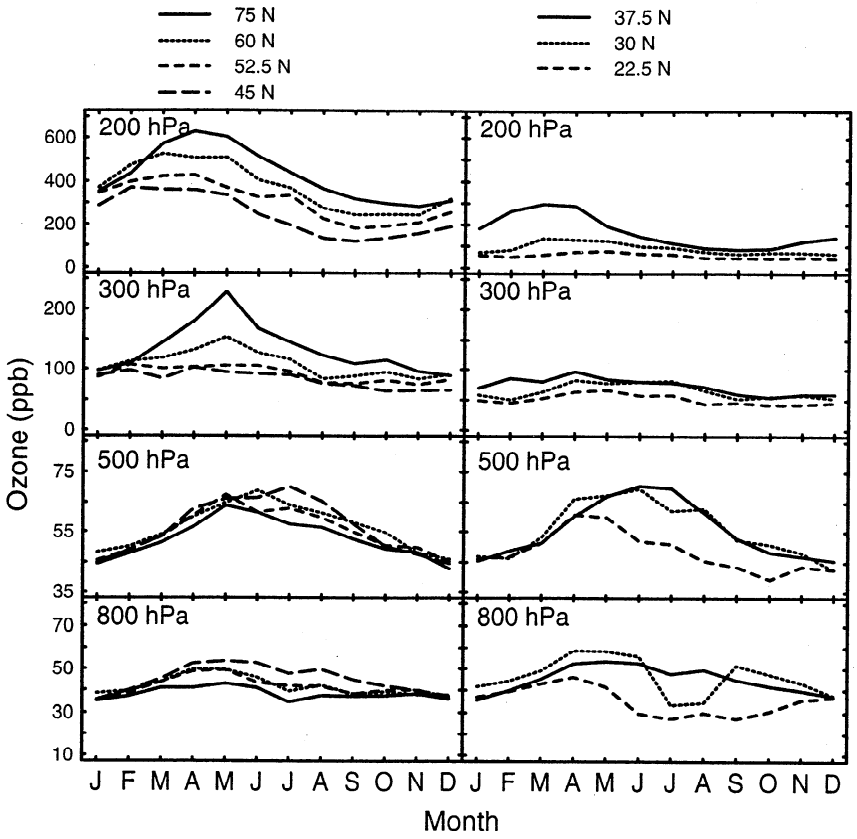


Figure 27. Annual cycle of ozone for 22.5° to 75°N in the climatology.

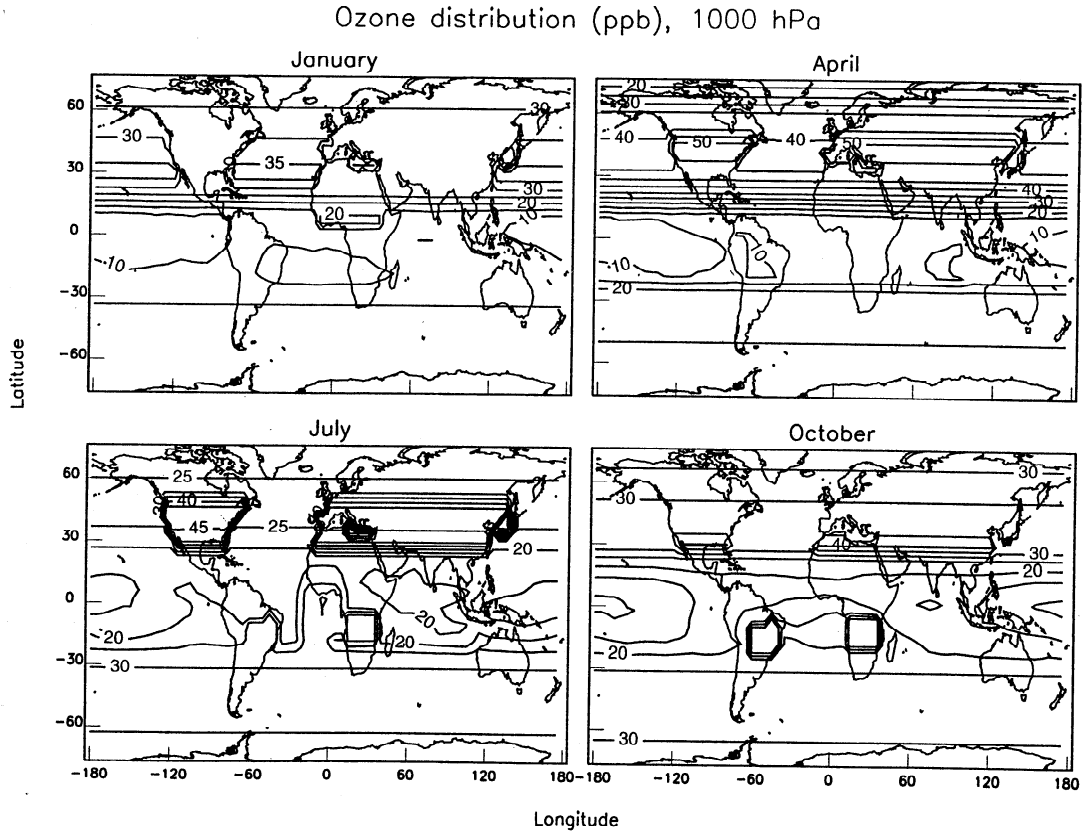


Figure 28a. Spatial distribution of ozone for the midseason months for 1000 hPa. Contours are given every 5 ppb.

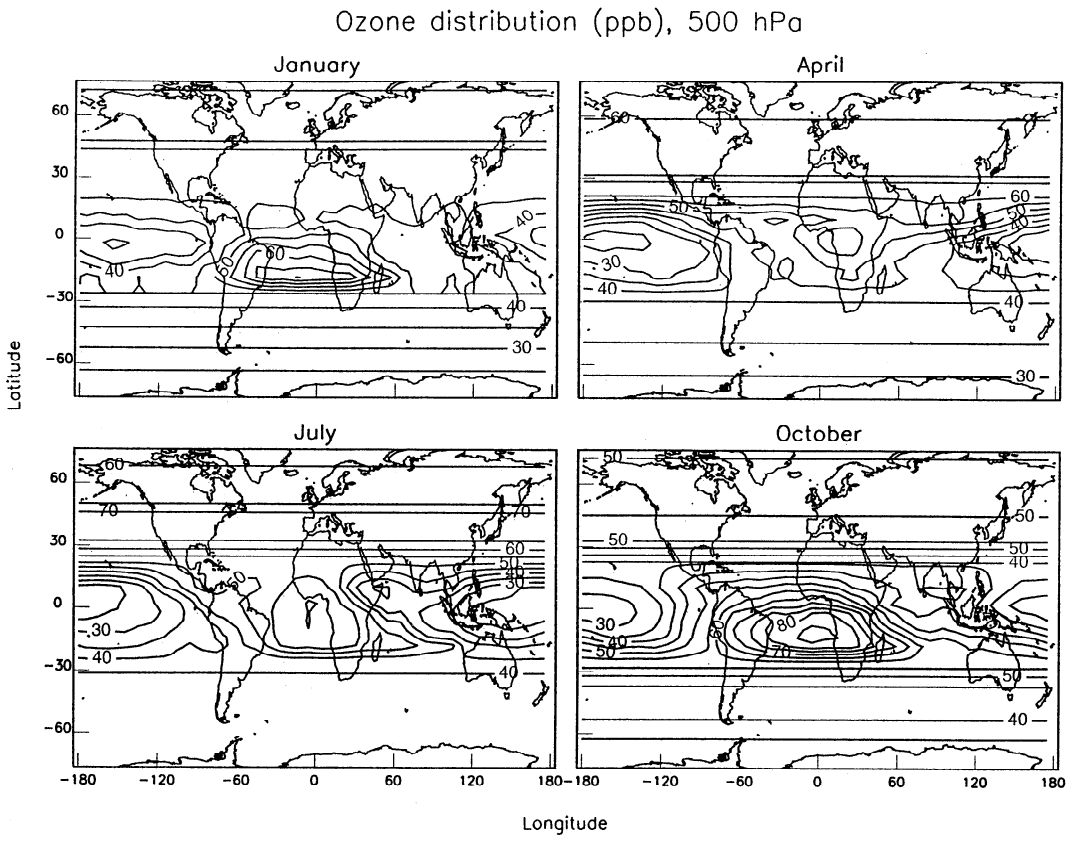


Figure 28b. Spatial distribution of ozone for the midseason months for 500 hPa. Contours are given every 5 ppb.

common period, 1980-1993, where possible. Data for shorter periods obtained since 1980 should not be influenced strongly by trends [*Logan,* 1994]. Measurements made with GDR sondes, which appear to be biased high in the troposphere, are not recommended for model evaluation. Data obtained prior to 1980 in southern Europe (Biscarosse, 44°N, and Cagliari, 39°N) extend the latitude range of the European sites; there is no evidence of a significant increase in ozone in southern France since 1980 based on measurements presented by *Beekmann et al.* [1994b] and *Ancellet and Beekmann* [1995].

Almost all the measurements in the northern tropics and subtropics shown here were obtained prior to 1980, and there is no information on trends in ozone for this latitude zone except for surface data from Hawaii, which shows a small increase in ozone in winter and spring prior to 1983 [*Oltmans et al.,* 1998]. Although the number of profiles is limited for Kennedy and Grand Turk, they provide a useful test of the ability of models to capture the different annual cycle of ozone over the Atlantic versus the mid-Pacific. These data should be supplemented for model evaluation with spring and summer data for Bermuda and the Azores [*Oltmans et al.,* 1996a]. The data from Panama in July 1977 provide good statistics for summer and are in agreement with the sparse data from the 1960s; the latter provide insufficient data to define monthly profiles. The Indian data are not recommended for model evaluation because of questions about data quality combined with sparse measurements. All the stations from the Southern Hemisphere are useful for model evaluation, but caution is required in using data for months with only a few measurements (see Table 2). In addition to the data given in Table 1, measurements from September and October 1992 from Brazil and southern Africa [*Kirchhoff et al.,* 1996; *Diab et al.,* 1996] provide further tests of models for the biomass burning season.

Monthly mean values of ozone are well defined for most middle- and high-latitude stations, with 95% confidence intervals of 6-15% for 800-500 hPa and 10-30% for 300-150 hPa for months with $N>20$ (see Figure 2). This is not the case for many of the subtropical and tropical stations, as shown in Figure 2 and Table 2. I recommend that the 95% confidence intervals be included in comparisons of model results with subtropical and tropical data for stations/months when $N<20$. The standard deviations of the monthly means should provide a good measure of the spread in the ozone distribution for $N>20$, but this is not the case for much of the tropical data.

Although the "long-term" (14 years) means are well defined, there is sufficient interannual variability in ozone that the mean in a given month and year will not necessarily fall within two standard errors of the long-term mean, as shown in Figure 30 for Hohenpeissenberg. This should be considered when comparing the averages given here with results of any model driven by assimilated meteorological data, or a model driven by 1 year of output from a general circulation model, which will not necessarily represent long-term average meteorology. Nevertheless, a model driven by meteorology that is not anomalous should be able to reproduce the general patterns seen in the ozone distribution, which are summarized in the concluding section.

I recommend that the data in Table 1, with the exceptions described above, be used to evaluate models of tropospheric ozone. *Wang et al.* [1998b], for example, evaluated the ability of their 3-D chemical tracer model (CTM) to reproduce the vertical distribution of ozone and the annual cycle of ozone in the lower,

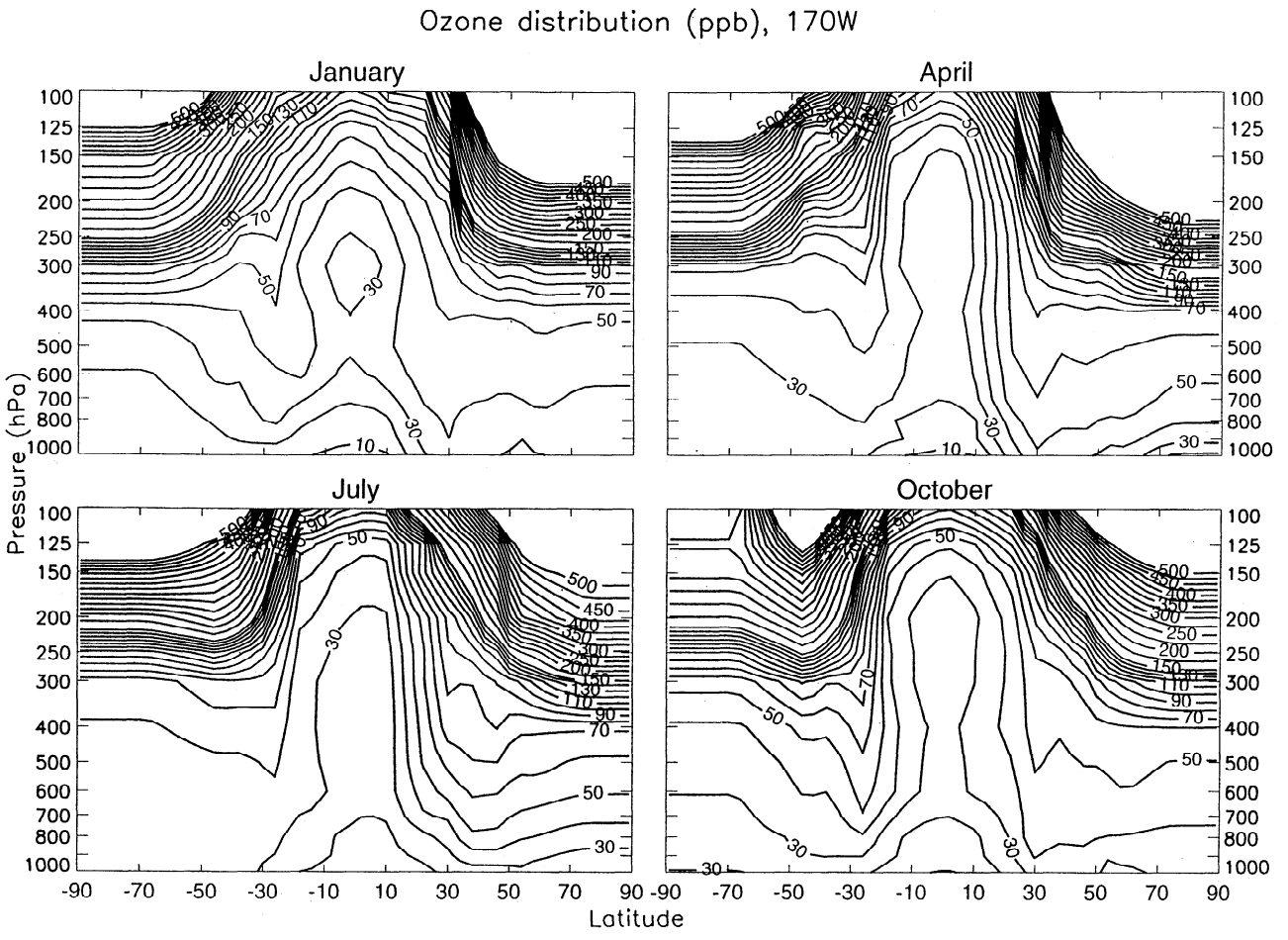


Figure 29a. Ozone climatology versus pressure and latitude for 170°W. Contours are given every 10 ppb up to 150 ppb, then every 25 ppb up to 500 ppb.

middle, and upper troposphere, represented by 800, 500, 300, and 200 hPa. A similar evaluation of an updated version of the IMAGES model [*Muller and Brasseur,* 1995] and of a preliminary version of the model of *Wang et al.* [1998b] was made as part of the Global Modeling Initiative [*Friedl,* 1997]. The model of *Wang et al.* [1998b] reproduces the annual cycle of ozone in the middle and lower troposphere of the extratropics, but it underestimates the vertical gradient in tropical ozone, implying that the the model has excessive vertical mixing across the trade wind inversion. IMAGES has about the correct annual mean amount of ozone at northern midlatitudes, but the model underestimates summer values, while it overestimates winter values, with an annual maximum that is 2-3 months too early. However, this model reproduces the summer maximum at European surface sites. Both models underpredict the spring maximum in ozone in the south tropical Atlantic (Natal and Ascension Island) by about 25 ppb [*Friedl,* 1997; *Wang et al.,* 1998b]. Hauglustaine et al. [1998] evaluated their 3-D model, MOZART, with data from 12 of the stations shown here; however, they did not show model results for Natal or Ascension Island.

Most three-dimensional tropospheric models have been evaluated with only a subset of the data shown here. *Muller and Brasseur* [1995], *Roelofs and Lelieveld* [1995], *Roelofs et al.* [1997], and *Berntsen and Isaksen* [1997] compared their model simulations with surface measurements, primarily those given by *Oltmans and Levy* [1994], and with some of the seasonal vertical profiles for eight stations presented by *Oltmans et al.* [1989a] and *Komhyr et al.* [1994]; *Crutzen and Zimmerman* [1991] showed no comparisons with ozone data, while *Levy et al.* [1985] relied on sonde data from *Logan* [1985] and the early years of surface data from the sites described by *Oltmans and Levy* [1994]. Of these studies, only the last used sonde data to test the ability of the model to predict the seasonal variation in the middle troposphere.

Some of the present generation of CTMs have fairly coarse vertical resolution, and the tropopause is not necessarily in the correct location in the extratropics [e.g., *Kasibhatla et al.,* 1996; *Wang et al.,* 1998b]. The tropopause heights of the sonde data are provided, as well as the annual cycle of ozone with respect to the tropopause, to provide a supplemental test of how well the model simulates the change in the behavior of ozone from the upper troposphere to the lower stratosphere at middle and high latitudes. A discussion of the use of the sonde data for testing models developed for stratospheric ozone is given by *Logan* [this issue].

Surface data provide an essential complement to the sonde data for model evaluation. As noted above, data from remote sites provided by *Oltmans and Levy* [1994] (see Figure 15) have been used to evaluate several 3-D CTMs. I recommend that data from the other sites in Figure 15 are used also, including the shipboard data for the Atlantic compiled by *Winkler et al.* [1988]. There is little diurnal variation in surface ozone at these oceanic and remote sites, while there is a significant diurnal variation in ozone for the rural sites in North America shown in Figure 16. For these

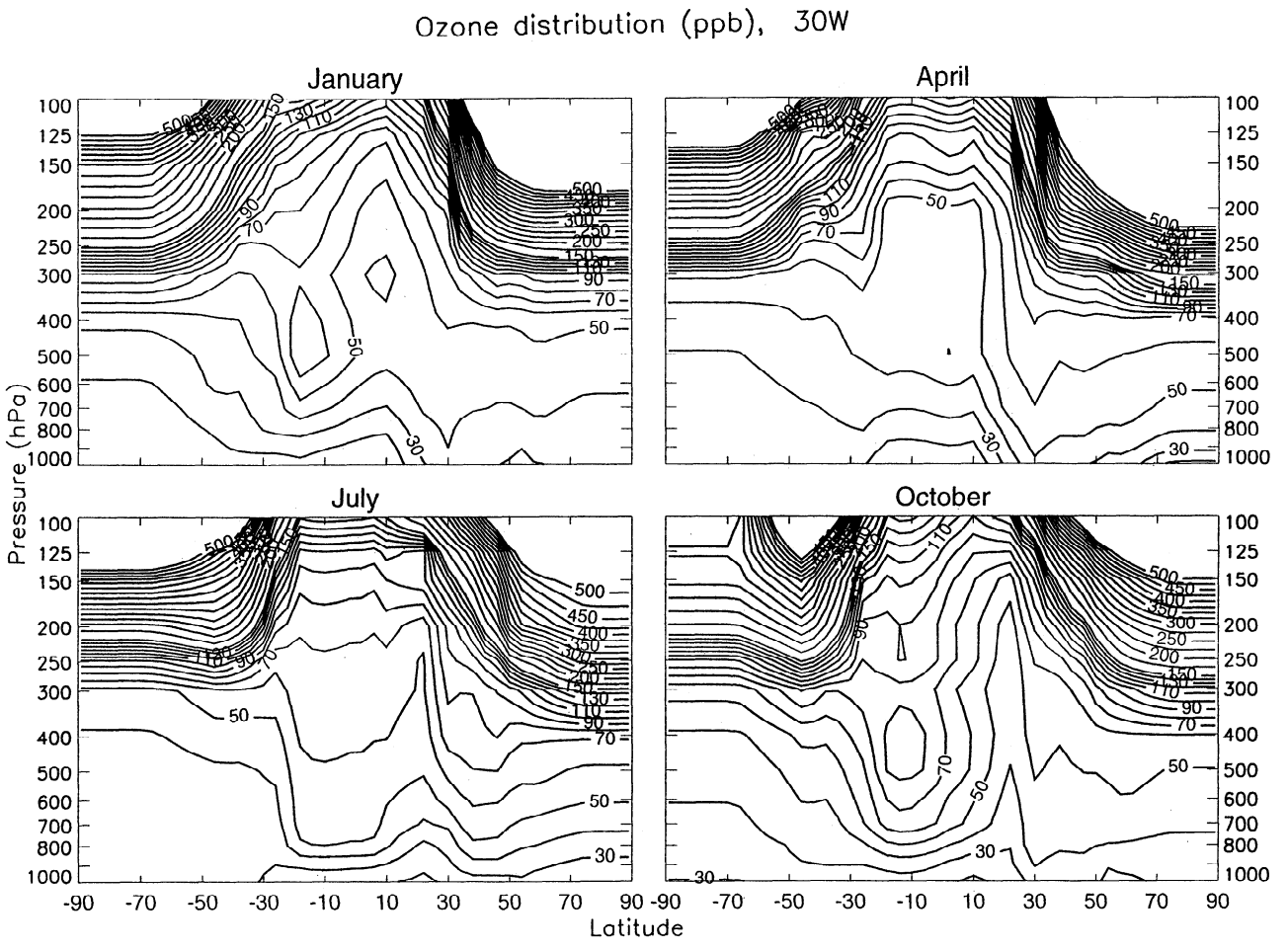


Figure 29b. Same as Figure 28a for 30°W.

sites the mean values of ozone from 1200 to 1600 LT provide a good representation of ozone values after the breakup of the nighttime inversion (see *Logan* [1988] for the diurnal variation of ozone for these sites). The 1200-1600 values have been used to test CTMs [e.g., *Jacob et al.,* 1993b; *Kasibhatla et al.,* 1996]. Data from the air pollution network in the United States are also recommended for model evaluation; these data provide a valuable test of the ability of models to reproduce enhancements in ozone over polluted regions [e.g., *Horowitz et al.,* 1998; *Liang et al.,* 1998]. A recent analysis of data from rural European sites should also find application for testing models [*Scheel et al.,* 1997].

In addition to the data presented in this paper, shipboard measurements of ozone [e.g,, *Piotrowicz et al.,* 1991], vertical profiles taken from ships in the Atlantic [*Smit et al.,* 1989; *Weller et al.,* 1996] and Pacific [*Kley et al.,* 1996], and aircraft data [e.g., *Marenco and Said,* 1989; *Marenco et al.,* 1990; *Browell et al.,* 1996] provide further information regarding the ozone distribution. The advantage of the sonde and surface site data for evaluation of 3-D CTMs is that they provide multiyear statistics as opposed to a snapshot of ozone concentrations. The data from campaigns are useful for testing CTMs driven by assimilated meteorological data.

Two years of ozone measurements obtained from regularly scheduled aircraft have recently been made available as gridded maps of the spatial distribution in the upper troposphere-lower stratosphere, and these data are being used to evaluate CTMs [*Thouret et al.,* 1998a; *Law et al.,* 1998; *Hauglustaine et al.,* 1998]. The MOZAIC data will become increasingly useful as the measurements and their analysis continues, as the geographic extent of the program expands, and as the data are dynamically stratified into tropospheric and stratospheric distributions. Similar comments apply to lidar data.

The climatology of ozone described in section 5 provides a further test of 3-D CTMs, in particular the ability of the model to simulate photochemical loss of ozone. Loss of ozone is driven primarily by HO_x chemistry and is almost independent of NO_x, depending primarily on H_2O, CO, and the ozone column. Computation of the global loss rate for ozone based on observation (the climatology) may be compared with the loss rate based on the model's ozone, providing a test of one component of the tropospheric ozone budget [e.g, *Wang et al.,* 1998b].

6. Conclusions and Recommendations

The ozone data shown here provide an essential test of 3-D chemical transport models in many regions of the world, with the exception of the northern tropics. These data may be used to test the ability of models to reproduce the vertical profile of ozone and the annual cycle in the lower, middle, and upper troposphere. Mean values, standard deviations, and the number of measurements for each station are available electronically (http://www-as.harvard.edu). For many stations, data are provided giving the altitude of the tropopause as well as the ozone distribution with respect to the tropopause, so that the influence of the position of

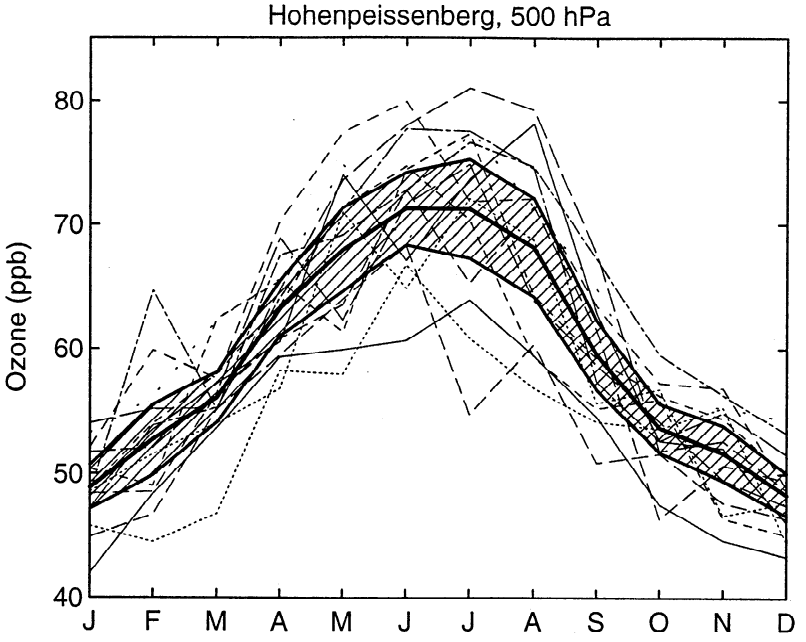


Figure 30. Annual cycle of ozone at Hohenpeissenberg at 500 hPa for 1980 to 1993. The thick solid line shows monthly means derived from 14 years of individual monthly means, and the cross hatched area shows 2 standard errors for interannual variability. The thin lines show means for individual means.

the model tropopause can be factored into the evaluation. The change in the phase of the annual cycle of extratropical ozone from a summer maximum at the tropopause and below to a spring maximum within 2 km above the tropopause, combined with the steep vertical gradient in ozone immediately above the tropopause, suggests that high vertical resolution (~1 km) will be required in models to simulate the ozone distribution near the tropopause.

The climatology provided here may be used to derive empirically based estimates of the global distribution of OH and of the chemical sink for ozone. It may be used also in climate models, to provide a priori profiles for satellite retrievals, and as initial conditions for 3-D chemistry-transport models. I do not recommend that 3-D model calculations of ozone be evaluated with the gridded climatology, because of the errors discussed above, but rather with the station data that were used to compile the climatology.

The distribution of tropospheric ozone is characterized best for middle and high latitudes of both hemispheres, although data are lacking for major regions such as continental Asia, South America, and the oceans. The lack of data from southern midlatitudes is not a serious problem given the apparent zonal symmetry in ozone there. Features that a model should be able to capture for these latitudes include (1) the broad midyear maximum in the Northern Hemisphere which extends from the middle troposphere to the tropopause, and the winter minimum; (2) the larger variability in summer than in winter; (3) the summer maximum in the lower troposphere over the polluted continents and the spring maximum over the remote continents and oceans; these features are apparent in the differences in boundary layer ozone over Europe and Canada near 50°N; (4) the winter maximum and summer minimum in the lower troposphere at southern middle and high latitudes and the spring maximum in the middle troposphere at midlatitudes; (5) the uniformity of ozone from 30° to 75° in both hemispheres in winter; (6) the maximum in ozone from 35° to 50°N in summer; (7) the interhemispheric gradient in ozone; (8) regional characteristics influenced strongly by the circulation, such as the low summer values caused by tropical airflow in the lower troposphere at Bermuda and the more southerly Japanese stations.

There is a dearth of contemporary data for ozone in the northern tropics and subtropics, and the climatology is most uncertain there. Consequently, there are few tests of model performance. Measurements of the latitudinal gradient in ozone in the northern hemisphere were last made three decades ago [*Hering and Borden,* 1967], and these data remain the only source of year-round data from the equator to 38°N in the western hemisphere. The older data show a summer maximum in midtropospheric ozone over the Atlantic near 20°N, while the recent data from the Pacific show a summer minimum and spring maximum, with the differences related to the origin of the air masses as described above. This provides one clear test for models. The old data suggest also that ozone may be as low over Panama as over Samoa in the same months, another issue that can be explored in models.

There is significant biomass burning in the northern tropics primarily from November to March in North Africa, central and South America (north of the equator), and southeast Asia (*Arino and Melinotte* [1995]; see fire maps at http://shark1.esrin.esa.it), but the consequences for ozone are not well documented. Contemporary ozone measurements are lacking for America and Asia, although MOZAIC measurements are now being made over Asia [*Thouret,* 1998]. Aircraft data show that ozone values of 60 to over 100 ppb are found over west Africa (1-4 km, 0°-10°N) in December to March, with lower values, 50-60 ppb above 4 km [*Marenco et al.,* 1990]. The ozone values for 1-4 km are as high as those found over southern Africa during the burning season [*Browell et al.,* 1996], while those above are lower than those found at Natal, Ascension, and Brazzaville. It is likely that much of equatorial North Africa is affected by elevated ozone, given the extent of fires across much of the continent, but the fate of the ozone formed in the boundary layer is not known. The tropospheric residual data imply that the influence of biomass burning on ozone is not so widespread nor as intense as that seen over the

south tropical Atlantic. In fact, the residual shows higher values over west Africa in the southern burning season (July to November) rather than at the time of local fires, but these values have not been verified by in situ measurements. The smaller signal in the residual may indeed be caused by a lower ozone column in the troposphere but may be influenced also by the different shape of the ozone profile, with higher values below 4 km than above. The residual may reflect the lack of sensitivity of the TOMS measurement to ozone in the lowest part of the troposphere [e.g., *Hudson et al.,* 1995]. The shape of the a priori ozone profile used in the TOMS retrieval [*Wellemeyer et al.,* 1997] underestimates ozone in the lower troposphere in biomass burning regions, and this may cause errors in the retrieval. There is clearly a need for in situ data to unravel the effects of biomass burning on ozone in the northern tropics. Reliable data for the vertical distribution of ozone are needed from the Caribbean, Africa, India, Asia, and the Pacific. There is a sonde program in India, but the publically available data are inadequate to define the ozone distribution, as discussed above.

The situation in the southern tropics is somewhat better, in that recent data are available from several locations in the Atlantic and Pacific, and measurements are currently being made at Natal, Samoa, Tahiti, Fiji, the Galapagos, Ascension Island, Indonesia, and Kenya (S. Oltmans and A. Thompson, personal communication, 1998). The current data provide excellent tests of the ability of models to capture the zonal asymmetry in ozone, and the combined influences of biomass burning and dynamics that cause the high ozone values over the south tropical Atlantic. They allow also better definition of the climatology than in the northern tropics.

The sonde data show that ozone is higher over the Atlantic compared to the western Pacific (Samoa) all year from about 800 hPa to the tropopause. Ozone is most similar in May and June, the months with minimum burning in the tropics. The seasonal cycle at Samoa in the middle and upper troposphere reflects the effect of biomass burning, with a peak 1 month later than over Natal. A secondary peak in December to March at Natal, Ascension, and Brazzaville appears to be caused by biomass burning in the northern tropics. Recent analysis of sonde and aircraft data from September 1996 confirms the influence of biomass burning over much of the tropical and subtropical Pacific in that season [*Schultz et al.,* 1998; S. J. Oltmans et al., manuscript in preparation, 1999; J. A. Logan et al., manuscript in preparation, 1999]. Two weeks of data from the equatorial western Pacific show that ozone can be exceedingly low in the upper troposphere (from zero to 25 ppb), a consequence of deep convection [*Kley et al.,* 1996]; these measurements are from the region of the tropical minimum in the residual maps. Preliminary data from Indonesia (7°S) show uniform ozone profiles of about 25 ppb in December to March, and higher values, with ozone exceeding 50 ppb in layers, in September to October; biomass burning is likely the cause of the elevated concentrations [*Komala et al.,* 1996]. There is a need for better characterization of ozone in the southern tropics also, particularly over Indonesia and the equatorial Pacific, and within South America and Africa. Data are lacking also for most of the southern subtropics, where longitudinal gradients are not well characterized by in situ data.

Analysis of standard errors in the tropical and subtropical data shows that at least 20 measurements are needed, likely over a period of a few years, to derive reliable mean values for the vertical profile of ozone. There have been suggestions of making two years of 1-3 measurements per week to increase current knowledge of the ozone distribution in the tropics. It is clear from existing data that thrice weekly measurements over 2 years would be a minimum set to define mean ozone profiles adequately; only 2 years of data would risk the selection of anomalous years and would not be an optimum strategy. Ozone is clearly influenced by biomass burning in many tropical locations, and there is evidence from CO observations of dramatic effects of year-to-year variations in spatial patterns of burning on trace gas distributions [*Reichle et al.,* 1990; *Connors et al.,* 1999].

Global measurements of the vertical profile of tropospheric ozone will be made for the first time from the Tropospheric Emission Spectrometer (TES), part of the Earth Observing System's CHEM platform scheduled for launch in 2002. TES will provide information on 3-4 levels in the troposphere, with vertical resolution of ~5 km in the nadir and 2.3 km in the limb [*Clough et al.,* 1995]. This will provide invaluable information of spatial patterns in ozone that cannot be obtained from in situ data. Ozonesonde measurements have started recently in Kenya (WOUDC web site), tropical Pacific sites (see above), Easter Island (R. Newell, personal communication, 1996), Surinam (6°N) [P. Fortuin, personal communication, 1998], and Ascension Island has recently reopened (A. Thompson, personal communication, 1997). These will be valuable additions to existing programs but do not fill all the gaps discussed above, particularly in the northern tropics. It is vital that ozone measurements are made available to the research community through WOUDC in a timely manner, which is the case for many but certainly not for all existing stations. Satellite measurements providing global information with high spatial resolution and in-situ measurements with high vertical resolution will be required to fully characterize the distribution of tropospheric ozone.

Acknowledgments. I would like to thank Amy Alberts and Inna Megretskaia for their able management and manipulation of the sonde data. I benefited from useful discussions with P. Kasibhatla and C. Spivakovsky. I thank all who made the thousands of soundings used in this study. The primary source of sonde data was the World Ozone and Ultraviolet Data Centre (http://www.tor.ec.gc.ca/woudc). Many thanks are due to those who provided their data to the Centre and to Edward Hare who has made WOUDC 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. J. Fishman and V. Brackett provided the residual data, and I had useful discussions about its reliability with JF. This work was funded with support from the National Air and Space Administration, grants NAG5-2688, NAGW-2632, and NAG1-1909 and the National Science Foundation, grant ATM-9320778.

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(Received August 10, 1998; revised December 4, 1998;
accepted December 10, 1998.)