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 variation at all in the stratosphere, the seasonal
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 strong enough to drive a total ozone column variation of
 about 5%, which is about one half the size of the
 variation observed in the Natal Dobson spectrophotometer
 data. The ozone concentration at Natal increases with
 height between the surface and about 500 mbar, almost
 linearly, from about 15 parts per billion by volume
 (ppbv) to about 38 ppbv, in autumn. For the spring
 average the ozone concentration increases from about 25
 ppbv at the surface to about 66 ppbv at 500 mbar. The
 sonde data suggest that limitations in aneroid pressure
 sensors used until 1986 caused the Natal sondes to
 indicate too much ozone above 6 mbar. Because of the

relative sparsity and uneven distribution in time of the ozone soundings, the data are not adequate to study ozone trends. The Dobson data time series shows no definitive ozone trend but displays a pronounced quai-biennial oscillation in ozone.

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OZONE CLIMATOLOGY AT NATAL, BRAZIL, FROM IN SITU OZONESONDE DATA

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ABSTRACT. Results are presented from analysis of a large ozone profile data set obtained from balloon ozonesonde soundings made at Natal, Brazil (6°S , 35°W) during the last 10 years (1978-1988). The measurements have been made through an Instituto Nacional de Pesquisas Espaciais (INPE)/NASA long-term collaboration program. The balloons were released by the Brazilian Air Force at the Natal rocket range. The data set is sufficiently large to provide useful climatology on the average ozone concentration behavior and its seasonal variation. The day-to-day ozone concentration variability in the troposphere is rather large, giving standard deviations of about 30-40% for seasonal averages. Maximum ozone concentrations occur during local spring, September-October, and minimum concentrations during late autumn, April-May. The seasonal variation in the troposphere is much larger than in the stratosphere. If there were no seasonal variation at all in the stratosphere, the seasonal variation observed in the troposphere alone would be strong enough to drive a total ozone column variation of about 5%, which is about one half the size of the variation observed in the Natal Dobson spectrophotometer data. The ozone concentration at Natal increases with height between the surface and about 500 mbar, almost linearly, from about 15 parts per billion by volume (ppbv) to about 38 ppbv, in autumn. For the spring average the ozone concentration increases from about 25 ppbv at the surface to about 66 ppbv at 500 mbar. The sonde data suggest that limitations in aneroid pressure sensors used until 1986 caused the Natal sondes to indicate too much ozone above 6 mbar. Because of the relative sparsity and uneven distribution in time of the ozone soundings, the data are not adequate to study ozone trends. The Dobson data time series shows no definitive ozone trend but displays a pronounced quasi-biennial oscillation in ozone.

Introduction

The vertical distribution of ozone has been measured at Natal (6°S , 35°W), in the troposphere and stratosphere, since November 1978, by the

Instituto Nacional de Pesquisas Espaciais (INPE), Brazil, in collaboration with the National Aeronautics and Space Administration, (NASA), U.S. Previous results were described by Kirchhoff et al. [1981], and with a focus on the stratosphere by Kirchhoff et al. [1983]. There are very few ozone sounding stations in the southern hemisphere. On the basis of data obtained from the World Ozone Data Center, Logan 1985 has listed 25 northern hemisphere stations compared with only four in the southern hemisphere, Natal being the only tropical station maintaining continuous measurements.

For the northern hemisphere, most of the long-term balloon sonde ozone profile data have been obtained at Payerge (47°N , 7°E) and Hohenpeissenberg (48°N , 11°E), but also from Resolute (75°N , 95°W), Goose Bay (53°N , 60°W), Wallops Island (38°N , 76°W), Tateno (36°N , 140°E), and Kagoshima (32°N , 131°E). Most of the soundings at southern mid-latitude come from Aspendale (38°S , 145°E). With its present data base, Natal has become the most important tropical sounding station whose data can be used to improve the a priori data base used for the solar backscatter ultraviolet (SBUV) satellite ozone profile retrieval algorithm.

It is usual to attribute larger tropospheric ozone concentrations to the northern hemisphere compared with concentrations in the southern hemisphere [Fishman et al., 1979]. Nevertheless, by comparing ozone sounding data from a station in Panama (Fort Sherman, 9°N , 80°W) with similar data obtained at Natal (6°S , 35°W), it was shown that larger tropospheric O_3 concentrations are actually observed at Natal [Kirchhoff, 1984]. In fact, surface ozone concentrations at Natal during local spring (September) are as large as values seen at Cape Grim (41°S , 145°E , Australia). In the middle troposphere, at 700 to 400 mbar, concentrations are comparable to values observed in the U.S. and Europe [Logan and Kirchhoff, 1986].

It was further pointed out that the seasonal variation of tropospheric ozone at Natal was sufficiently strong to influence the variation seen by instruments that measure the total ozone column. These early findings stimulated continuation of the sounding program at Natal.

The measurement instrument that has been used is the electrochemical concentration cell (ECC) ozonesonde. This ozonesonde is a small, balloon-borne device developed at the National Oceanic

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and Atmospheric Administration (NOAA) [Komhyr, 1969; Komhyr and Harris, 1971]. The ozone sensor of the ECC ozonesonde is an iodine/iodide redox concentration cell, composed of two platinum electrodes immersed in neutral buffered iodide solutions of different concentrations in the anode and cathode chambers. An electric current is generated when air containing ozone is pumped into the cathode. Along with measurements of ambient air pressure, temperature, and instrument temperature, this current is converted into ozone partial pressure (in nanobars), the primary data product for the ECC ozonesonde. The ECC sondes in this data set used 1.5% potassium iodide cathode sensing solution. Since 1982 the sampling pump efficiencies for the sondes [Torres, 1981] were individually measured before flight; also, the ozonesondes were calibrated in the laboratory against UV ozone photometers, whose calibrations were traceable to the photometric ozone standard of the National Institute of Standards and Technology (U.S.). The calibrations were single measurements of ozone, at an ozone partial pressure of about 120 nbar, by the ECC and the UV instruments. Ratios of these measurements provided correction factors for the sondes.

ECC ozonesondes are carried aloft on balloons, accompanied by standard meteorological radiosondes that measure temperature and pressure from the ground to the burst altitude of the balloon. In special cases the ECC sondes were also tracked by radar throughout their flights. The C band tracking radar at Natal has an estimated measurement accuracy of 30 m at a range of 50 km.

This paper presents ozone results for Natal obtained during the last 10 years. On the basis of this data set we derive an ozone climatology for the equatorial troposphere and stratosphere in the region of Natal to 34 km altitude. Some aspects of the data are also compared with results from other ozone measurement techniques (SBUV, Dobson). Finally, long-term ozone variations are briefly discussed.

Results

The long-term ozone profile measurement program at Natal was started in late 1978 when six soundings using ECC sondes were made in November and December. Including results for 1988, a total of 179 ozone soundings have been made. This represents a considerable data base, the largest for an equatorial site. The data were, however, not always obtained from soundings uniformly distributed with time. Difficulties in maintaining a continuous supply of ozonesondes caused interruptions in the sounding schedule. Table 1 shows the number of sondes released during different periods of time. Note that the number of soundings per year ranged from 32 in 1984 to 7 in 1987.

Figure 1 shows a histogram of the number of soundings made during different time periods of the day. The majority of sondes were released around 0900 local standard time (LST), or 1200 UT. The reason for this concentration of releases around a specific time of day is that most soundings were synchronized with satellite overpasses of the SBUV series. Surface ozone

TABLE 1. Number of Soundings in Different Time Periods, at Natal, During 1978 to 1988

Period	Number of Soundings
Year*	
1978*	6
1979	19
1980	17
1981	15
1982	12
1983	15
1984	32
1985	29
1986	16
1987	7
1988	11
total	179
Month	
Jan.	22
Feb.	12
March	13
April	18
May	10
June	9
July	11
Aug.	15
Sept.	17
Oct.	20
Nov.	21
Dec.	11
During periods ¹ of ozone maxima ¹	
1978-1980	14
1981-1982	5
1983-1984	24
1985-1986	9
1987-1988	6
During periods ² of ozone minima ²	
1978-1980	11
1981-1982	4
1983-1984	8
1985-1986	15
1987-1988	3

* The data distribution in each year is shown in Figure 2.

¹ Sept., Oct., Nov.

² March, April, May.

measurements at Natal [Kirchhoff, 1989] show that the diurnal ozone concentration variation at this site is rather small. Model calculations [Herman, 1979] show only very small diurnal changes in stratospheric O_3 below 40 km. The sonde results may therefore be considered representative for any daytime period both in the troposphere and stratosphere.

Launches of the ECC ozonesondes, including the radiosondes, were made using 2000 g balloons. Table 2 lists statistics on the heights attained. Soundings at Natal with these balloons have reached apparent heights of 39 km, or 3 mbar as

Fig. 1. Histogram of the number of ozone soundings made at Natal versus times of launch.

indicated by the radiosonde pressure sensors. Errors in these pressure indications and how they affect the ozone data are discussed later. All sondes reached a height of at least 18.6 km; slightly less than 50% of the sondes reached 32.5 km altitude, and only 5% of the sondes reached 39.2 km.

Data Quality and Quality Criteria

A Dobson spectrophotometer has also been operated at Natal since the start of the sounding program. The option of whether normalizing ECC ozonesonde data to the Dobson instrument total ozone content is appropriate or not has been discussed previously [Kirchhoff et al., 1983; Logan and Kirchhoff, 1986]. It has been shown by Logan and Kirchhoff [1986] that the results are not sensitive to the use of the Dobson scaling procedure. We chose not to normalize our data set to the Dobson integrals, but we used normalization factors as exclusion parameters for

TABLE 2. Maximum Balloon-Burst Height Statistics

Percentage of Sondes	Pressure, mbar	Height, km
100	70.0	18.6
99	60.0	9.5
98	50.0	20.6
97	40.0	22.0
96	35.0	22.8
94	30.0	23.8
89	25.0	24.9
85	20.0	26.4
79	17.5	27.2
78	15.0	28.3
71	12.5	29.5
64	10.0	31.0
47	8.0	32.5
38	7.0	33.4
24	6.0	34.4
18	5.0	35.7
10	4.0	37.2
6	3.5	38.1
5	3.0	39.2

Shown are the percentages of soundings that reached the indicated heights (above sea level). See text about accuracies of the altitude sensors of the radiosondes.

defining unreliable sonde data. All ECC soundings whose O_3 integrals fell below 0.8 or above 1.2 of the Dobson instrument values were excluded from the analyses reported here. On average, the agreement between the sonde and the Dobson integrals was good as shown in Table 3 by the ECC/Dobson total ozone ratios, for the years 1978 to 1988. The ratios range from 1.07 to 0.95, except in 1980 when the ratio was 1.12. Using

TABLE 3. Total Ozone Ratios Obtained From ECC and Dobson Instruments, and Ozone Concentration Ratios From ECC and UV Absorption Instruments Obtained at Natal During 1978-1988

Year	Number of Soundings	Total Ozone Ratio ECC/Dobson	Number of Measurements	O_3 Concentration Ratios ECC/UV
1978	6	1.07 $\pm$ 0.19*		-
1979	15	1.05 $\pm$ 0.12		-
1980	12	1.12 $\pm$ 0.04		-
1981	11	1.06 $\pm$ 0.08		-
1982	9	1.01 $\pm$ 0.19	6	1.06 $\pm$ 0.21
1983	12	0.99 $\pm$ 0.10	15	1.04 $\pm$ 0.05
1984	27	1.02 $\pm$ 0.13		-
1985	22	0.95 $\pm$ 0.08		-
1986	9	0.95 $\pm$ 0.09	16	1.09 $\pm$ 0.05
1987	6	1.01 $\pm$ 0.04	5	1.09 $\pm$ 0.04
1988	7		11	1.06 $\pm$ 0.07

* Indicated variabilities are standard deviations. Note that number of soundings here are actually the number of events when both ECC and Dobson data were available.

this quality control procedure, about 5% of the soundings were not included in further analyses.

A second data quality indicator resulted from prelaunch ECC instrument calibrations with an ultraviolet absorption ozone meter, used routinely after 1981. Several companies manufacture these instruments, for example, Dasibi, Monitor Labs, Thermo Electron, etc. For simplicity, we will designate these ozone meters as UV. The average ECC/UV meter ozone ratios and

their standard deviations are also shown in Table 3. For a total of 133 soundings made simultaneously with Dobson observations, the sondes give a mean total ozone value of 277 DU (Dobson units), whereas the Dobson instrument mean is 278 DU, with standard deviations of 18 and 13 DU, respectively. There have been indications by several tests that in some height ranges the sondes overestimate ozone, while in others they may underestimate ozone. However, this comparison between the total ozone content of the two instruments is excellent. [Barnes et al. 1985] have made laboratory tests on ozonesondes of the same type used at Natal and found that the sondes may overestimate ozone in the troposphere by about 8 to 14%. [Hilsenrath et al. 1986] have also made tests on different types of ozonesondes flown simultaneously and report similar results. Other performance characteristics of the sondes have been discussed by Torres and Bandy [1978]. Although balloon ozone soundings may not be perfect, they are presently the only method available for the long-term insitu measurement of ozone in the troposphere and stratosphere at any site.

Ratios of total column ozone measured by the ECC and Dobson instruments are plotted in Figure 2. The plots present information also on the temporal distribution of the available data. Large data gaps occur in 1982 and 1983, and again in 1987 and 1988.

Average Ozone

The average tropospheric ozone profile for Natal is displayed in Figure 3. It shows the general pattern of ozone distribution in the troposphere with the following characteristics: in the region between the surface and 500 mbar (5.9 km) the ozone mixing ratio profile maintains a rather constant rate of increase with height, of 5.5 ppbv/km. Above 500 mbar and up to 200 mbar (12.4 km) the mixing ratio is roughly constant, at about 52 ppbv, and above 200 mbar, the mixing ratio gradient slopes strongly towards the much larger mixing ratios of the lower stratosphere. This general behavior of the mixing ratio variation with height is a characteristic that can be observed as well in most individual profiles obtained at Natal. It

Fig. 2. Total ozone measured with ECC and Dobson instruments shown in terms of ratios for 1978-1988.

Fig. 3. The average tropospheric ozone profile using all data 1978-1988. The horizontal bar shows typical size of the standard deviations from the average.

TABLE 4. Tropospheric Ozone Averages Obtained From 157 ECC Ozonesondes Launched at Natal in the Period 1978-1988

Pressure, mbar	Height,* km	Ozone, nbar	Standard Deviation %	Ozone, ppbv	Ozone, 10^{11}cm^{-3}	Temperature, K
1000	0.1	19.9	52.8	19.9	4.7	300.1
900	1.0	20.6	43.2	22.9	5.1	291.4
850	1.5	21.9	38.4	25.7	5.4	288.9
800	2.0	25.1	39.0	31.4	6.3	287.2
700	3.2	28.5	39.6	40.7	7.2	282.1
600	4.4	27.3	36.3	45.5	7.1	275.0
500	5.9	26.3	34.2	52.7	7.1	267.3
400	7.6	21.3	33.3	53.2	5.9	256.5
350	8.6	19.6	33.2	55.9	5.6	249.1
300	9.7	14.8	39.9	49.3	4.4	241.0
250	10.9	14.1	35.5	56.3	4.4	230.7
200	12.4	10.7	43.0	53.5	3.5	218.8
150	14.2	11.4	34.2	76.2	4.0	205.4
125	15.3	11.4	33.3	91.5	4.1	199.4
100	16.5	12.9	34.1	129.1	4.7	195.2

A plot of the tropospheric ozone profile in ppbv is shown in Figure 3.

* Above sea level.

is also characteristic of other tropical sites [Kirchhoff et al., 1988].

Average ozone concentrations at specific pressure altitudes between 1000 and 100 mbar, derived from 157 ECC ozonesonde profiles at Natal are listed in Table 4. Averages for March-April-May and September-October-November are shown in Tables 5 and 6, respectively.

The average ozone profile in the stratosphere, derived from 157 ECC ozone soundings made at Natal during 1978-1988, is shown in Figure 4. Average ozone concentrations, at specific pressure altitudes between 70 and 3 mbar are given in Table 7. The dashed curve above 6 mbar in Figure 4 was obtained from a series of optical ozonesondes launched at Natal on rockets in 1985 [Barnes et al., 1987]. Above

34 km there is considerable disagreement between the ozonesonde data and the rocket results. The rocket measurements show lower ozone amounts and a significant difference in the ozone scale height. The shape of the rocket profile above 34 km agrees closely with concurrent SAGE II measurements, both at the equator and at northern mid-latitudes [Cunnold et al., 1989]. In addition, the ECC ozone data also diverge from the MAP Interim Reference Ozone Model [Keating and Young, 1985] and from the first-year SBUV ozone measurements [McPeters et al., 1984] with the ECC sonde data showing significantly higher ozone mixing ratios at 5 to 3 mbar. We feel that the problem above 34 km is in the ECC ozonesonde data presented here.

Previously published comparisons of ozone

TABLE 5. Tropospheric Ozone Averages Obtained From 40 Soundings During March, April and May at Natal During the Period 1978-1988

Pressure, mbar	Height,* km	Ozone, nbar	Standard Deviation %	Ozone, ppbv	Ozone, 10^{11}cm^{-3}	Temperature, K
1000	0.1	15.1	35.8	15.1	3.6	299.9
900	1.0	15.7	25.5	17.4	3.9	292.2
850	1.5	16.1	27.3	19.0	4.0	289.5
800	2.0	17.4	27.6	21.8	4.4	287.2
700	3.1	19.3	29.5	27.6	4.9	282.0
600	4.3	18.7	34.8	31.2	4.9	275.0
500	5.8	19.1	34.6	38.2	5.1	267.9
400	7.5	15.5	32.9	38.8	4.4	257.1
350	8.5	14.4	38.2	41.1	4.1	249.9
300	9.6	11.6	41.4	38.7	3.5	241.8
250	10.9	10.0	33.0	40.1	3.1	231.5
200	12.3	8.1	42.0	40.4	2.7	219.0
150	14.1	9.8	43.9	65.2	3.4	204.9
125	15.2	10.1	42.6	81.1	3.7	198.7
100	16.5	11.2	35.7	111.8	4.2	193.5

* Above sea level.

TABLE 6. Tropospheric Ozone Averages Obtained From 58 Soundings During September, October and November, at Natal During the Period 1978-1988

Pressure, mbar	Height,* km	Ozone, nbar	Standard Deviation %	Ozone, ppbv	Ozone, 10^{11}cm^{-3}	Temperature, K
1000	0.3	25.3	57.3	25.3	6.1	300.2
900	1.2	25.4	43.3	28.2	6.3	290.8
850	1.7	27.1	32.5	31.9	6.8	288.4
800	2.2	32.0	29.4	40.0	8.0	287.2
700	3.3	37.5	29.3	53.6	9.6	282.6
600	4.5	34.3	27.7	57.1	9.0	275.0
500	6.0	32.9	26.1	65.7	8.9	267.2
400	7.7	26.3	31.2	65.7	7.4	256.6
350	8.7	24.3	33.3	69.3	7.0	249.0
300	9.8	18.3	42.1	60.9	5.5	240.9
250	11.0	18.4	34.8	73.7	5.8	230.7
200	12.5	12.9	39.5	64.7	4.3	218.8
150	14.3	13.0	33.1	86.5	4.6	205.6
125	15.4	12.6	33.3	101.1	4.6	199.0
100	16.7	13.7	32.1	136.7	5.0	195.6

* Above sea level.

measurements using different techniques [Barnes et al., 1985; Hilsenrath et al., 1986] have shown that the ozone values from ECC ozonesondes tend to be too low at pressure altitudes higher than about 10 mbar. In the present study, on the contrary, the ozone values above 6 mbar are higher (in comparison to other techniques mentioned). It will be shown next that the high ozone values indicated by the ECC ozonesondes at Natal at pressure altitudes above 6 mbar (34 km) are not a problem of the ozone sensors themselves, but result from errors in the aneroid pressure sensors of the meteorological radiosondes used in conjunction with the ozonesondes.

Fig. 4. The average stratospheric ozone profile using all Natal ECC data, 1978-1988. The horizontal bar represents the standard deviation from the average. Above 34 km, the dash circle line indicates ozone results obtained from optical rocket sondes at Natal. See text for discussion.

The radiosonde aneroid pressure sensors are set at ground level at pressures near 1000 mbar. For these individual pressure settings, errors of a few millibars can be expected. They only represent small errors at ground level but become significant at high altitudes.

For a large set of radiosondes there occurs a distribution of pressure readings both higher and lower than the actual pressure. This distribution of pressures about the mean extends over most of the pressure range of the sonde measurements. For altitudes up to 30 km there is no sign of systematic problems with the pressure sensors [Parsons et al., 1984]. However, as the balloons reach their upper altitude limit and burst, a natural selection process occurs that removes the instruments with higher pressure outputs from the data set. This results from the fact that relatively few balloons reach high altitudes.

This effect of a skew toward lowpressure readings at the upper altitude limit for the ozonesondes is more evident in the calculation of ozone mixing ratios (the ratio of the ozone partial pressure to atmospheric pressure). The erroneously low atmospheric pressure readings give ozone mixing ratios that are too large. Table 7 shows ozone mixing ratios at 3 mbar that are larger than the mixing ratios at 4 mbar.

The problem with the pressure sensors is not evident from smaller series of just a few instruments or even a few tens of instruments. The fraction of balloons that make it to the highestpressure altitudes is small. A large data set, such as the one presented here is necessary to assess the effects of this problem. We believe that this problem exists in all ozone data sets from balloon-borne instruments using aneroid pressure sensors to give altitude information. The problem can be minimized by using hypsometer pressure sensors in the radiosondes. The hypsometers give greatly improved pressure measurements at pressure altitudes above about 20 mbar [Parsons et al.,

TABLE 7. Stratospheric Ozone Averages Obtained From 157 ECC Ozonesondes Launched at Natal in the Period 1978-1988

Pressure, mbar	Number of Soundings	Height,* km	Ozone nbar	Standard Deviation %	Mixing ratio ppmv	Ozone, 1012cm ⁻³	Temperature, K
70.0	157	18.6	30.0	27.3	0.43	1.07	200.7
60.0	156	19.5	46.8	20.7	0.78	1.64	204.9
50.0	154	20.6	65.3	18.2	1.31	2.26	208.3
40.0	152	22.0	91.3	13.8	2.28	3.10	212.5
35.0	150	22.8	106.0	13.9	3.03	3.55	215.0
30.0	147	23.8	119.1	13.4	3.97	3.94	217.6
25.0	140	24.9	134.0	11.7	5.36	4.38	220.7
20.0	134	26.3	140.7	10.0	7.04	4.53	224.0
17.5	124	27.2	138.9	9.1	7.94	4.43	226.1
15.0	123	28.2	132.6	10.0	8.84	4.19	228.4
12.5	111	29.5	118.1	12.4	9.45	3.70	230.5
10.0	101	31.0	97.7	17.4	9.77	3.03	233.1
8.0	74	32.5	77.2	20.9	9.65	2.37	235.4
7.0	59	33.4	68.2	24.0	9.74	2.08	236.6
6.0	37	34.4	62.0	25.0	10.33	1.89	237.3
5.0	29	35.7	54.7	26.9	10.94	1.65	239.7
4.0	16	37.2	47.8	29.7	11.95	1.44	240.6
3.5	10	38.1	48.5	34.8	13.86	1.46	241.0
3.0	8	39.2	43.1	42.7	14.37	1.30	242.1

A plot of the stratospheric ozone profile in cm⁻³ is shown in Figure 4. Ozone data above 5 mbar are invalid due to erroneous pressure measurements by the radiosonde instruments flown with the ECC sondes.

* Above sea level

1984]. Hypsometers have been used with all ECC ozonesondes flown at Natal since 1986. In addition, balloon altitudes can be measured with tracking radars. ECC instruments flown from Natal have been radar tracked in special cases.

Pressure sensor errors impose an upper altitude limit for which ozone data presented here are valid. It is our judgement that this limit is 6 mbar, or about 34 km.

Seasonal Variations

The seasonal variation in tropospheric ozone between the surface and 200 mbar is shown in Figure 5. There is nearly a factor of 2 difference between minimum and maximum values, a systematic feature observed every year. It is shown in Figure 5 that in the period of high ozone values, August to November (local spring), the tropospheric abundance measured from the surface to 200 mbar is 37.3 DU (October), which represents 13.5% of the total ozone column of 277 DU. In the period of low ozone values, February to May (local autumn), the ozone content of the troposphere between the surface and 200 mbar is 21.0 DU (in March for example), 7.6% of the average total ozone column of 277 DU. These results indicate that Dobson measurements at Natal are influenced less by tropospheric ozone during autumn than in spring [Logan and Kirchhoff, 1986].

The seasonal cycle of ozone is strong throughout the troposphere, with the variation slowly decreasing with height, as shown in Figure 6. Near the ground or in terms of the tropospheric ozone integral, the variation

between minimum and maximum is roughly a factor of 2 (Figures 5 and 6). In the whole ozone column this variation is only about 10% (Figure 8).

There is considerable difference in the vertical ozone profiles between spring and autumn as shown in Figure 7. In spring, 60 ppbv ozone are common in the free troposphere of Natal. This is not to be expected on the basis only of the relative movement of the subsolar point with

Fig. 5. Average seasonal variation in ozone column amount at Natal between the surface and 200 mbar. The vertical bars are the standard deviations, and the numbers above the months represent the number of sonde data sets comparing the averages.

Fig. 6. Average seasonal ozone variations at selected heights in the troposphere and stratosphere.

respect to Natal. Essentially, no difference in tropospheric ozone should exist at times of the two equinoxes, when solar intensity is largest on an annual basis to a first approximation. The Natal maximum during spring, centered in September/October, may be the result of photochemical ozone production triggered by biomass burning products transported to Natal either from central Brazil or from Africa, as discussed by Kirchhoff and Nobre [1986]. Pollution products from both possible sources would be injected into the Natal troposphere after traveling for large distances. However,

Fig. 7. Average vertical O_3 profiles in the troposphere for equinox periods. Curve A, for fall, was obtained from 40 soundings during March, April, and May. Curve B, for spring, was obtained from 58 soundings made during September, October, and November.

since the seasonal variation at Natal is slightly larger at the surface, the distant African contribution seems to be the dominating one, with combustion gases being transported to Natal by prevailing easterlies.

The Dobson instrument observations at Natal (Figure 8) indicate for the whole data set a seasonal variation in total ozone of only 5% with respect to the yearly average of 277 DU. The standard deviations of the Dobson instrument total ozone monthly means are of the order of 3%-5%, whereas for the ECC total ozone averages the sigmas are about twice as large (Table 8). Figure 8 also shows the relative seasonal variations for tropospheric and stratospheric ozone. The triangles denote Dobson spectrophotometer measurements, which are the result of averaging daily observations. The

Fig. 8. Average seasonal relative variation in total ozone at Natal as measured with a Dobson spectrophotometer (open triangles) and ECC ozonesondes (closed circles). The relative variation of tropospheric ozone between the surface and 200 mbar (open squares) is also shown, for comparison.

TABLE 8. Monthly Mean Total Ozone Amounts Measured at Natal With ECC Ozonesondes and With a Dobson Spectrophotometer During 1978-1988

Month	Total O ₃ ECC DU	Total O ₃ Dobson, DU	Total O ₃ K + Y DU	Tropospheric O ₃ ECC, DU
Jan.	268 ± 43.3	275 ± 11.8	257	27.9 ± 3.9
Feb.	261 ± 22.5	269 ± 8.0	256	22.2 ± 1.9
March	266 ± 20.4	268 ± 6.5	259	21.0 ± 3.1
April	267 ± 19.7	264 ± 4.9	260	20.9 ± 4.4
May	252 ± 22.4	260 ± 11.6	260	18.3 ± 4.4
June	256 ± 18.3	267 ± 12.1	261	23.6 ± 4.0
July	289 ± 33.5	274 ± 9.9	264	29.8 ± 5.4
Aug.	283 ± 33.2	283 ± 9.1	270	32.9 ± 6.0
Sept.	272 ± 27.6	290 ± 10.3	275	36.2 ± 8.1
Oct.	287 ± 22.4	289 ± 8.6	267	37.3 ± 5.4
Nov.	291 ± 24.8	282 ± 8.3	266	34.3 ± 5.3
Dec.	308 ± 25.6	276 ± 11.5	260	35.4 ± 8.5
Average	275.0	274.7	262.9	28.3
σ	16.6	9.7	5.6	6.9

Also shown are monthly mean total ozone values in the troposphere between the surface and 200 mbar, determined from ECC ozonesonde observations. For comparison, monthly mean total ozone amounts are shown for the Nimbus 7 TOMS data of Keating and Young [1985].

* Averages ± standard deviations are shown.

relative variation in the troposphere is much stronger and is shown in Figure 8 for comparison (actual values taken from Table 8).

Comparison With Other Data and Long-Term Variations

Dobson spectrophotometer and ECC ozonesonde total ozone data for 1978-1988 are compared with Keating and Young [1985] monthly mean ozone averages from NIMBUS 7 total ozone mapping spectrometer (TOMS) satellite ozone observations. These are 4-year averages for the time period November 1978 to September 1982. The horizontal

resolution of the TOMS observations is 50 km, and the monthly averages were derived from data for 5° latitude belts.

The TOMS data show lower ozone values of about 259 DU from January to June, with higher values of about 268 DU for the rest of the year. The Dobson monthly mean values are 12 DU larger than the TOMS values. One reason for the discrepancy in the data from the two kinds of instruments is the averaging of data over large areas that is made with the TOMS values, resulting in a general smoothing process. It has been shown elsewhere that the TOMS instrument ozone values are generally about 6% lower than Dobson instrument ozone values [Barthia et al., 1984]. It has also been pointed out by Klenk et al. [1985] that the Dobson network currently uses absorption coefficients that were adopted in 1968, and that the implementation of the new NIST (former NBS) ozone absorption coefficients would lower Dobson direct sun ozone column amounts by about 3.5%, which would improve comparisons.

The large variabilities associated with the ECC ozonesonde average monthly total ozone values (Table 8) preclude the possibility of obtaining meaningful long term ozone trends from the ECC data. The Dobson instrument data, however, are examined for total ozone trends. Following Oltmans and Komhyr [1986], the seasonal cycles have been removed from the Dobson data set by subtracting from the monthly means of each year corresponding monthly averages, for the entire data period. The results, sometimes called residuals, are shown in Figure 9, smoothed by a 12 month running mean. They indicate a very clear oscillation with period of about 2 years. Hilsenrath and Schlesinger [1981] obtain similar wave amplitudes using Nimbus 4 UV data. This QBO

Fig. 9. Twelve-month running means of total ozone content residuals (monthly means minus the climatological monthly means) from Natal Dobson data showing clear evidence for a quasi-biennial oscillation (QBO) in ozone.

(quasi biennial oscillation) has been described before, for example, by Quiroz [1981] and Hilsenrath and Schlesinger [1981], but has not been shown previously specifically for the Natal Dobson data. A linear regression line fit to this data set indicates a small decrease of ozone of $(-0.03 \pm 0.06)\%$ per year, which is, however, not statistically significant at the 95% confidence level.

Summary

A 10-year ozonesonde database is presented for the low-latitude station Natal (6°S , 35°W). Although data are presented for pressure altitudes of up to 3 mbar, they are considered unreliable above 6 mbar due to radiosonde pressure measurement errors.

Maximum concentrations of ozone at Natal occur in September/October, local spring. This maximum is about 5% higher than the annual average for the total ozone column. In the troposphere, however, between the surface and 200 mbar, the seasonal variation is much stronger. The tropospheric partial column of ozone is 37.3 DU in October, and 20.9 DU in March. The difference in these extremes is 16.4 DU or 5.9% of the total average annual ozone column. This shows that the seasonal variation in the troposphere accounts for about one half the annual cycle modulation of the entire ozone column. The seasonal variations are largest near the surface, with amplitudes decreasing slowly with height. In terms of average mixing ratios, values vary from 15.1 to 25.3 ppbv at the surface from autumn to spring, whereas at 500 mbar the variation is from 38.2 to 65.7 ppbv. It is noted that the Natal maximum occurs after the dry season periods of central Brazil and central Africa (July) which, in principle, would allow biomass burning products to be transported from the sources to Natal.

Long-term variations in ozone could not be detected from the ECC sonde data because of the relative sparsity and uneven distribution in time of the ozone soundings. Analysis of the Natal Dobson data showed no statistically significant trend in ozone. The data, however, exhibited a strong QBO signal with period of 27 months.

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