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Total ozone trends in the tropics

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Abstract. Measurements with Dobson spectrophotometers are used to study ozone trends at Cachoeira Paulista (22.7°S, 45.0°W) and Natal (5.8°S, 35.2°W) in Brazil. The time series are from 1974 to 1997 for Cachoeira Paulista and from 1978 to 1997 at Natal. Using a model that accounts for the quasi-biennial oscillation, seasonal variation, and solar cycle, a trend in total ozone of –2.0±1.2% per decade was found at Cachoeira Paulista for the period 1974 to 1997. At Natal the trend was –0.8±0.8% per decade over the period 1978 to 1997. The annual values of column ozone reported at Cachoeira Paulista are lower in the 1990's than in the previous two decades: total ozone amounts are 12.5 Dobson units (DU), or 4.6%, lower in the 1990s than those observed during the 1970s. From a monthly analysis it is shown that these ozone decreases have occurred in all months of the year. There was no statistically significant change between the 1980s and 1990s at Natal.

1. Introduction

Concerns about atmospheric ozone depletions, as a result of the catalytic action of nitrogen oxides from the exhaust of high-flying supersonic aircraft [*Crutzen*, 1971; *Johnston*, 1971] and the catalytic destruction resulting from human-produced chlorofluorcarbons [*Molina and Rowland*, 1974], were expressed in the early 1970s. Considering the general lack of atmospheric ozone observations in the Southern Hemisphere, the Brazilian National Institute for Space Research (INPE) started long-term observations of atmospheric ozone by installing a Dobson spectrophotometer (114) at Cachoeira Paulista (22.7^{o}S, 45.0^{o}W), São Paulo, Brazil, in May 1974. In addition, another Dobson spectrophotometer (093) became operational at Natal (5.8^{o}S, 35.2^{o}W), Rio Grande do Norte, Brazil, in November 1978. Both instruments are still operational and have provided the longest series of total ozone measurements in Brazil. *Sahai et al.* [1982] and *Kane et al.* [1985] have presented some of the earlier results on total ozone measurements at Cachoeira Paulista and Natal.

Influences of anthropogenic and natural causes on variations of atmospheric ozone (including long-term trends) have recently been studied using both satellite and ground-based observations (for global trends [*Chandra and Stolarski*, 1991; *Stolarski et al.*, 1992; *Reinsel et al.*, 1994; World Meteorological Organization (WMO), 1994; *Bojkov and Fioletov*, 1996; *Bojkov et al.*, 1995; *Jackman et al.*, 1996; *Harris et al.*, 1997; *Ziemke et al.*, 1997]; for more regional features [*Kalicharran et al.*, 1993; *Kundu and Jain*, 1993; *Chandra et al.*, 1996; *Tourpali et al.*, 1997; *Zerefos et al.*, 1997; *Kirchhoff et al.*, 1991, 1997]. This paper discusses time series of total ozone measurements obtained with Dobson spectrophotometers at Cachoeira Paulista (1974-1997) and Natal (1978-1997).

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2. Results and Discussion

The Dobson spectrophotometer 093 (hereinafter referred to as D093) was recalibrated in June 1994, during the World Meteorological Organization (WMO) intercomparison campaign at Izaña, Spain. In May 1995 an intercomparison between the Dobson spectrophotometer 114 (hereinafter referred to as D114) and D093 was carried out at Cachoeira Paulista. Several intercalibrations were made with the National Oceanic and Atmospheric Administration (Boulder) international standard Dobson (I-83) and traveling standard lamps. These data, deposited in the World Ozone Data Center in Toronto, are the result of a full re-evaluation (in consultation with some members of the WMO Ozone Assessment-98 panel) of the series obtained at the two stations, based on these and other previous calibrations and intercomparisons.

The Cachoeira Paulista series is almost continuous, but unfortunately, there are a few gaps in the observational series at Natal. Table 1 shows the periods of total ozone measurements made at the two stations. Only direct Sun observations from the Dobson have been used in the present study. All the ozone data presented have been calculated using the *Bass and Paur* [1985] ozone absorption cross sections.

Figure 1 shows plots of the monthly averages of total ozone measured at Cachoeira Paulista (1974-1997) and

Table 1. Details of Total Ozone Observations at Cachoeira Paulista and Natal

Station	Observations
Cachoeira Paulista	1. Observation period May/1974 to Dec./1997. 2. Months with no observations: Jan. 1993, Jan. 1996, and Jan. 1997.
Natal	1. Observation period Nov. 1978 to Dec. 1997. 2. Months with no observations: Nov. and Dec. 1980; April to June 1986, July to Dec. 1991, Jan. to July 1992, and June to Sep. 1994.

Natal (1978-1997). In order to obtain continuity, monthly means for the few missing months of Tabel 1 have been substituted with the Total Ozone Mapping Spectrometer (TOMS) overpass data obtained from the satellites Nimbus 7 (Version 7.0), Meteor 3 and Earthprobe. For Cachoeira Paulista, simultaneous observations for 210 months with TOMS overpasses and for Natal simultaneous observations for 190 months were available. Both Cachoeira Paulista and Natal show good agreement with TOMS data with correlation coefficients $r^2 = 0.83$ and $r^2 = 0.79$, respectively. Note that TOMS provides observations on nearly all days in a month, whereas the Dobson instruments are operated only on working days. The TOMS and the above Dobson station records do not show any systematic differences. The mean (and standard deviation) for the monthly differences between the Dobson and TOMS measurements at Cachoeira Paulista (210 months) is -1.00 ± 0.36 Dobson units (DU), whereas at Natal (190 months) it is -0.37 ± 0.32 DU. The thick lines in Figure 1 show the 12 points running means (to reduce the seasonal variation), and the horizontal lines indicate the whole period average ozone amounts at each station (Cachoeira Paulista 270.0±13, DU; Natal 265.5±10 DU). Note that this Natal average is smaller than the one reported by *Kirchhoff et al.* [1991], who analyzed the period 1978-1988 (average total ozone obtained from Electro Chemical Concentration ozonesonde measurements was 275.0±16.6 DU and 274.7±9.7 DU from Dobson observations), a consequence of the local downward ozone trend (see also discussion with Figure 2).

The monthly means of total ozone from the Dobson spectrophotometer data for both stations have been used to estimate long-term trends by a statistical model [*Bojkov et al.*, 1995; *Harris et al.*, 1997] that includes effects of the quasi-biennial oscillation (QBO) and solar cycle components. The results are presented in Table 2. In order to estimate the effect of missing months in the Dobson data, the data set has been completed with TOMS data, and

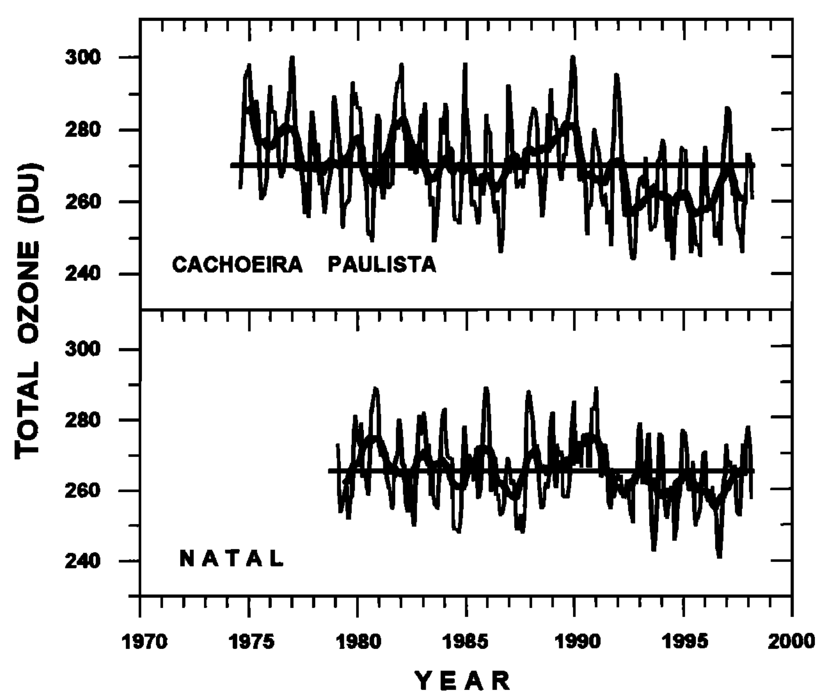

Figure 1. Monthly mean total ozone amounts (DU) observed at Cachoeira Paulista (1974-1997) and Natal (1978-1997). A few missing monthly means (Table 1) were substituted by TOMS data. The solid line is a 12 point running mean. The horizontal lines are the average total ozone for the period (Cachoeira Paulista, 270.0 DU; Natal, 265.5 DU).

Table 2. Total Ozone Trends for Different Periods and Data Sets

Station	Period	Summer DJF	Fall MAM	Winter JJA	Spring SON	Year
		Decline % per decade $\pm\sigma$				
Natal	Nov.1978 to Dec.1997	-0.2±1.3	0.4±1.3	-2.3±1.3	-1.1±1.2	-0.8±0.8
Natal*	Nov. 1978 to Dec.1997	-0.3±0.3	0.3±1.2	-2.3±1.3	-1.4±1.3	-0.9±0.8
Cachoeira Paulista	May 1974 to Dec.1997	-1.3±1.6	-1.5±1.7	-3.3±2.1	-1.9±1.6	-2.0±1.2
Cachoeira Paulista*	May 1974 to Dec. 1997	-1.6±1.7	-1.4±1.7	-3.4±2.1	-2.1±1.6	-2.2±1.2

*Dobson data + TOMS for missing months.

the trends recalculated. The Dobson + TOMS (for missing months) data set gives very similar trend results, also shown in Table 2. The effect of differences in the different TOMS instruments, and possible data biases of these with the Dobson instrument, have not been considered.

The total ozone trends at Cachoeira Paulista and Natal are –2.0±1.2% per decade and –0.8±0.8% per decade, respectively. These observed trends are comparable with those reported by *Bojkov and Fioletov* [1996], that is, about –2.0% per decade over the tropics, but not significant at the 2σ level (that is, 2σ is larger than the change). For a shorter period, *Stolarski et al.* [1992] presented results of trends over the tropics using TOMS data up to March 1991 and found near-zero trends in the tropics, and these were also not statistically significant.

Stronger ozone declines in the tropical region during the early 1990s (1990-1994) have been reported by *Tourpali et al.* [1997] (see also *Harris et al.* [1997] and have been attributed to the effects of the chlorine loading of the atmosphere. For this period, are larger declines also observed in the present data? Several factors may be responsible for changing the ozone behavior. For example, smaller ozone values observed in the period 1991-1993 could be associated with effects of Mount Pinatubo volcanic eruptions (June 1991), which resulted in a large increase in sulfate aerosol in the stratosphere throughout the globe [*WMO*, 1994]. About 2 weeks after the Mount Pinatubo eruptions, a strong enhancement in column SO_2 was observed over Brazil [*Sahai et al.*, 1997]. A few percent decrease in total ozone is generally attributed to the eruption of Mount Pinatubo [*Chandra*, 1993; *Randel and Cobb*, 1994]. After the Mount Pinatubo eruptions, *Harris et al.* [1997] have observed large negative ozone anomalies in the tropics of about 15 DU (6%) occurring near the equator from September to November 1991 and in the southern tropics in mid 1992. Some of the observed declines in the 1990s could also be associated with a stronger QBO [*Bojkov and Fioletov*, 1996], the influence of the solar cycle (1990 solar maximum and 1995-1996 solar minimum), and El Niño-Southern Oscillation (ENSO) (1992 and strong 1997-1998) influences. The overall effects of the QBO and the solar cycle terms on total ozone levels are about 0.2 to 0.3% per decade [*Chandra et al.*, 1996], whereas the ENSO signal in the total ozone is typically about 1% [*Harris et al.*, 1997]. *WMO* [1994] have reported that the observed middle- and high-latitude ozone losses and associated trends are largely due to anthropogenic chlorine and bromine compounds reacting with ozone, and that these losses increase with latitude. Thus larger ozone declines should result for Cachoeira Paulista than for Natal.

Wallace et al. [1997] have reported a general increase in HCl abundance by a factor between 3 and 4 from 1970 to 1990 from observations at Kitt Peak Observatory in Arizona. Also, *Harris et al.* [1997] have pointed out that the stratospheric levels of chlorine have increased in a nonlinear way by a factor of 5-6 in the last 30 years (bromine levels have also risen) and should have caused an acceleration in the total ozone trends, if indeed they are driven by halogen chemistry. Therefore the observed ozone declines during the period 1990-1997 could be the result of a combination of several geophysical phenomena. For the present data set at Cachoeira Paulista and Natal, the

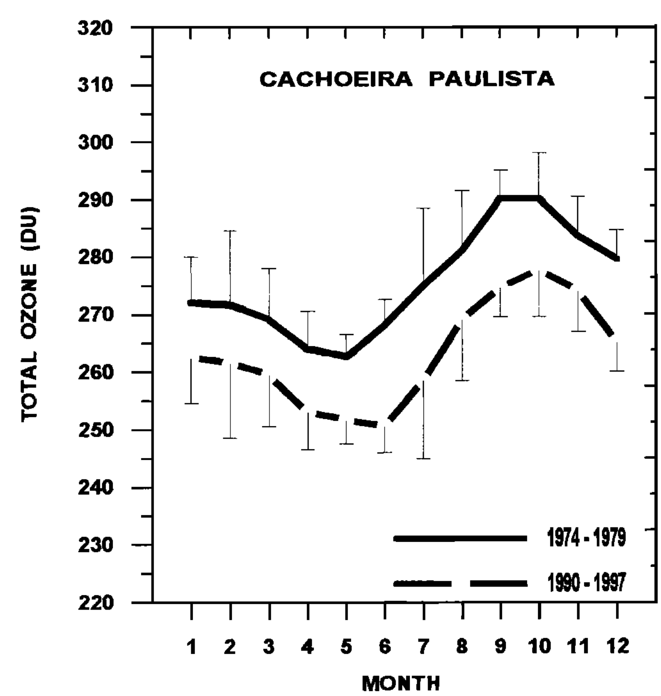

Figure 2. Mean seasonal variations observed (only Dobson data) at Cachoeira Paulista during 1974-1979 (continuous line) and 1990-1997 (dashed line). Also, the standard deviations for each month are shown by vertical bars.

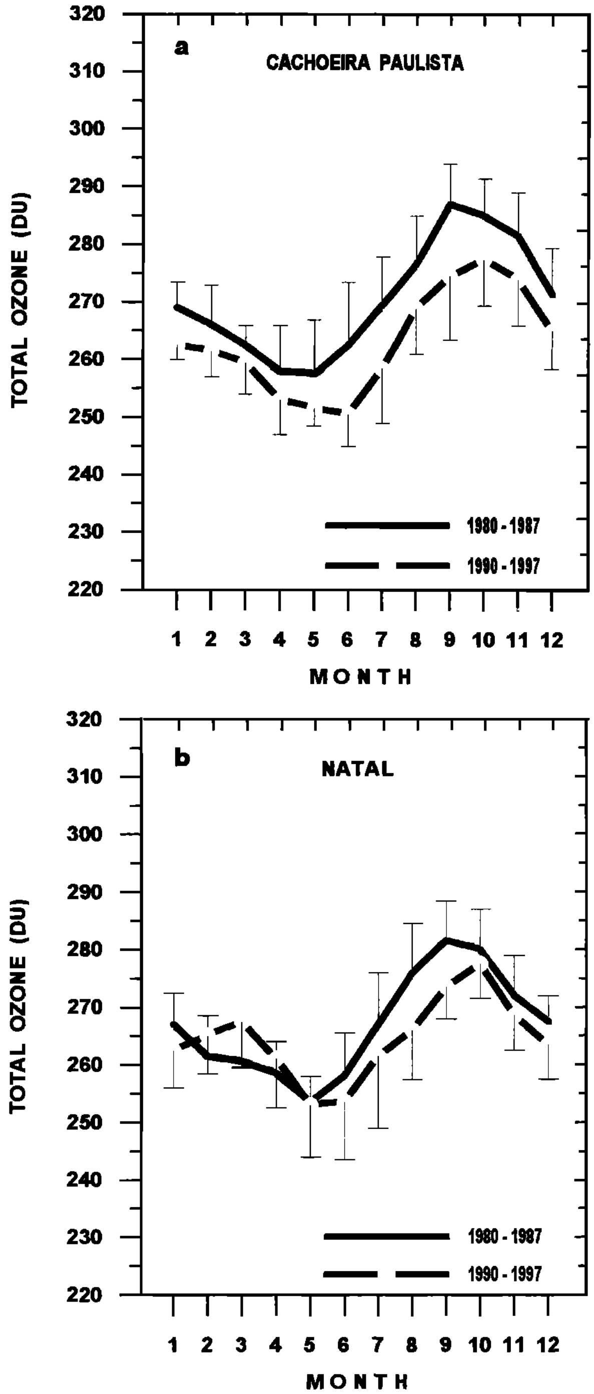

Figure 3. (a) Same as Figure 2, but for the period 1980-1997 and 1990-1997.
(b) - Same as Figure 2, but for Natal, and periods 1980-1987 and 1990-1997.

calculated "trends" for the shorter periods, for both stations, give results that are not statistically significant.

It is interesting to investigate if the ozone declines occur in a particular month of the year or if this is rather uniform. Figure 2 shows the mean seasonal variations observed at Cachoeira Paulista for the periods 1974-1979 (earlier period) and 1990-1997 (more recent period) for comparison. It is seen that total ozone shows considerable decreases during all months of the year in the 1990s compared with the period 1974-1979. The difference of the two seasonal variations (1974-1979 and 1990-1997) and the standard error of the difference is 12.5±3.8 DU, for Cachoeira Paulista. Figures 3a and 3b show similar plots for Cachoeira Paulista and Natal for 1980-1987 and 1990-1997, respectively. The differences with their standard errors for different periods of observations of ozone at Cachoeira Paulista and Natal are presented in Table 3. The difference between the 1970s and the 1990s for Cachoeria Paulista is statistically significant. However, at Natal and Cachoeira Paulista, for the comparison of the 1980s with the 1990s, this difference is statistically insignificant.

The ozone decreases observed at Cachoeira Paulista from the 1970s to the present would also mean a considerable change in the UVB radiation reaching the Earth's surface. Assuming that total ozone reductions of 1% typically cause an increase of about 1.2% in erythemally active UV irradiance [*McKenzie et al.*, 1991], the total ozone decline in the tropics during this period would have resulted in about a 5% increase in erythemal UVB irradiance. So far, there have been no reports on measured increases of this radiation in the tropics.

Figure 4 shows the overall mean seasonal variations (calculated from monthly averages) observed in total ozone at Cachoeira Paulista (1974-1997) and Natal (1978-1997). The mean seasonal patterns at Cachoeira Paulista and Natal are very similar. Both stations show a maximum in spring, as already shown by earlier results [*Sahai et al.*, 1982; *Kirchhoff et al.*, 1991]. The amplitude of this yearly variation is about 11%, that is, the difference between maximum and minimum, at both stations. The difference of the ozone averages between Cachoeira Paulista and Natal and the standard error of the difference is 4.0±3.4 DU. At Natal, part of the spring maximum has been associated to biomass burning [*Logan and Kirchhoff*, 1986; *Ziemke and Chandra*, 1998].

3. Conclusions

Long-term total ozone observations are presented for the Brazilian stations Natal (equatorial station,1978-1997) and Cachoeira Paulista (tropical station, 1974-1997). As a quality control measure, comparisons were made of the

Table 3. Ozone Average Differences for Different Periods of Observations (Only Dobson Measurements)

Station	Periods	Difference
Cachoeira Paulista	1974-1979 1990-1997	-12.5±3.8 DU
Cachoeira Paulista	1980-1987 1990-1997	-7.5±3.9 DU
Natal	1980-1987 1990-1997	-3.5±3.2 DU

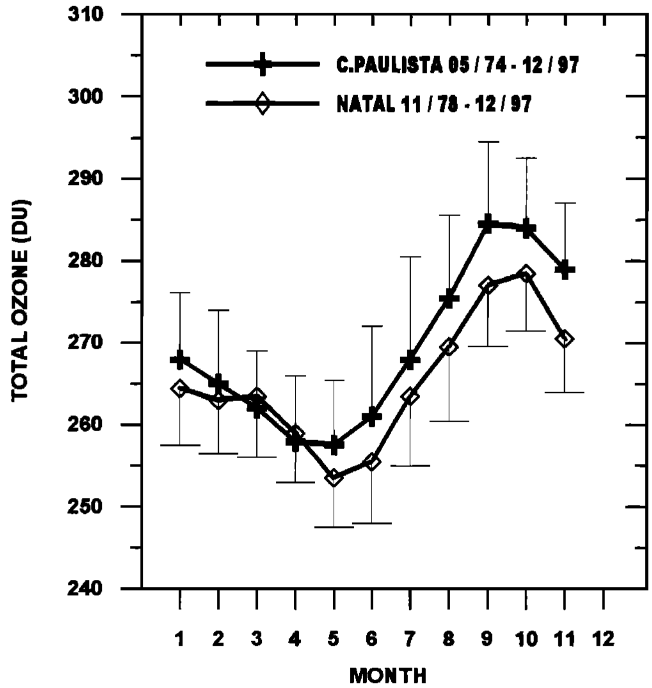

Figure 4. Mean seasonal variations of total ozone (only Dobson data) observed at Cachoeira Paulista (plus) and Natal (diamond) with the standard deviations for each month shown by vertical bars (one side only).

ground-based data with TOMS data; the difference for simultaneous data (210 monthly averages) was only -1.0 DU for Cachoeira Paulista, whereas it was -0.37 (190 monthly averages) for Natal. As shown in earlier analyses, both stations show a distinct seasonal variation (amplitude about 11%) with maximum in spring.

The long-term trends at both stations show expected decreases of total ozone. Using the whole time series of more than 20 years of Dobson data for Cachoeira Paulista, the total ozone trend (1974-1997) is –2.0±1.2% per decade, whereas at Natal (1978-1997) it is –0.8±0.8% per decade. A comparison of the annual means of total ozone observations at Cachoeira Paulista during the more recent 1990-1997 period with the earlier period of 1974-1979 shows that ozone has decreased by about 4.6% in all months of the year.

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