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AT NATAL (5.9°S, 35.2°W)

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ABSTRACT

Ozone density profiles obtained through ECC sonde measurements taken during the first three years of a joint INPE/NASA long term program of measurements at Natal have been analysed. Time variations, as expected, are small. Outstanding features of the data are tropospheric densities substantially higher than those measured at other stations, and also a total ozone content that is higher than the averages given by satellite measurements.

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## Introduction

The vertical distribution of ozone has been deduced since about 1930 from ground based observations of the attenuation of solar radiation at certain wavelengths using an inversion technique known as the Umkehr method. Reliable instruments capable of measuring ozone in direct soundings were developed only some 30 years later (Regener, 1960; Komhyr, 1969; Komhyr and Harris, 1971). Direct in situ soundings using detectors flown on balloons are presently made on a routine basis at several stations around the world, in an effort to accumulate ozone data over long periods for the study of possible long term changes of the ozone density. Instruments to detect ozone have also been launched on rockets (Krueger, 1973) and satellites (Prabhakara et al, 1971; Hilsenrath et al, 1979; Hilsenrath and Schlesinger, 1981). All these different techniques of measuring ozone complement each other. Probes on balloons are limited to heights of about 5 to 3 mbar, and thus the data for this region and above are mostly determined through the use of rockets. For the satellite data reduction, on the other hand, one must know, in general, an initial ozone density model profile for any given station in order to transform the radiance data into density data. Such ozone density model profiles have been derived based mainly on the availability of balloon-sonde data and rocket data (Hilsenrath and Dunn, 1979; Mateer et al, 1980).

Other methods of detecting ozone, mostly applied to the mesosphere, include millimeter wave observations (Penfield et al, 1976),

stellar occultation measurements (Riegler et al, 1976; Riegler et al, 1977; Hays and Roble, 1973) and an indirect mesospheric ozone determination through measurements of atmospheric sodium (Kirchhoff et al, 1981a).

The ozone measurement program at Natal is the result of a joint effort between NASA and INPE to obtain ozone data using both ECC sensors launched on balloons and optical sensors flown on rockets. A Dobson spectrophotometer (number 93) is also in operation at Natal since November 1978 in collaboration with NOAA. In a preliminary report (Kirchhoff et al, 1981b) it was shown how the average ozone profile at Natal, deduced from a two-year data set, differs from an average profile deduced from measurements at several stations between  $\pm 30^\circ$  latitude. The present study, covering a period of three years, discusses the ECC ozonesonde measurements at Natal in considerably more detail.

### Results and Discussion

ECC sondes of the 3A series have been launched from Natal ( $5.9^\circ$  S,  $35.2^\circ$  W) on balloons on 56 occasions, most of them around 12 UT. On three of these events the measured total ozone content compared to the Dobson value was unacceptably high (larger than the average plus three times the mean square error). This has been attributed to an as yet unidentified malfunction of the ECC sensor and therefore the data for these three days (April 13, 1979, sonde 560;

July, 5, 1980, sonde 905; and August 8, 1980, sonde 902) have been removed from the main data base, thus reduced to 53 profiles. On the basis of the results of Geraci and Luers (1978), who showed that the average total content of ozone determined by ECC sensors is within less than  $\pm 5\%$  of the Dobson value, we have not normalized our data base. The measurement program was started in November of 1978 for the launching of approximately one to two balloons each month. This analysis includes data until October 1981.

Figure 1a shows the yearly average profiles of ozone. The number of measurement events (balloon flights) for each year are shown in parenthesis, and the horizontal bars represent typical mean square errors. In the troposphere and lower stratosphere there seems to be little variation from the average year to year profile (less than about 20%). The troposphere shows a maximum in  $O_3$  density at about 700 mbar whereas the main  $O_3$  peak is at about 20 mbar. The largest variability can be seen above 10 mbar but one should remember that the data for this height region is subject to perhaps the largest errors for individual measurements, probably larger than the generally believed uncertainty of about  $\pm 5\%$  that one attributes to the normal ECC ozone sensor, (Torres and Bandy, 1977; Mani and Sreedharan, 1973) due to the decreasing pump efficiency of the device at high altitudes (Torres, 1981). On the other hand, few of the balloons have flown as high as 3 mbar. This level was reached only 3 times in 1981, 5 times in 1980 and 3 for the data of 1978-1979. The larger ozone



densities observed during 1980, especially close to the main ozone peak, may be associated to the biennial oscillation discussed by Hilsenrath and Schlesinger (1981). At about 30°S, Tolson (1981) and Ramanathan (1963) show the biennial maximum to occur at even years, but closer to the equator the phase seems to reverse.

It is interesting to note also that the larger ozone density has its peak at a slightly lower altitude. This characteristic will be seen again when comparing average ozone profiles during periods of very low and very high total ozone content. It seems to be a generally observed trend (London, 1978).

The ozone density at the ground (1000 mbar) also shows a rather large variability from day to day which can be seen by a large mean square error of about 50%. This value decreases toward higher altitudes being about 30% at 700 mbar.

The three year average ozone profile measured at Natal is shown in Figure 1b, where the horizontal bars represent again the mean square error. There is a secondary ozone peak in the troposphere at 3.1 km, with a density of  $7.5 \times 10^{11}$  molecules  $\text{cm}^{-3}$  (29.3 nbar). Minimum ozone density is  $4.0 \times 10^{11}$  molecules  $\text{cm}^{-3}$  (12.2 nbar) at 12.4 km (200 mbar), somewhat lower than the tropopause located at about 16.5 km (100 mbar). The main ozone peak has a density of  $4.5 \times 10^{12}$  molecules  $\text{cm}^{-3}$  (144.2 nbar) at 26.4 km (20 mbar). This Natal average profile is compared to an average result compiled for several other stations located within  $\pm 30^\circ$  latitude, according to results of

Hilsenrath and Dunn (1979), shown in Figure 1b by the dash-dotted curve labeled HD. The Natal average total ozone content is .287 cm STP and the HD curve shown is an average for .285 cm STP of ozone total content. It is clear from inspection of Figure 1b that the main ozone peak at Natal is slightly displaced upwards in comparison to the HD average. At the 700 mbar level the ozone densities at Natal are 30% higher than the HD average and the tropospheric structure is similar. The difference in tropospheric density is artificially enhanced in the interpolations of Hilsenrath and Dunn (1979) for .250 and .300 cm STP as can be seen in Kirchhoff et al. (1981b) where the Natal average in the troposphere is larger by almost 100%. It is interesting to note that the standard ozone profiles for low latitude derived by Mateer et al (1980) do not show the tropospheric secondary ozone peak, but give a tropospheric ozone partial pressure almost constant up to the tropopause.

Before one concludes that tropospheric ozone at Natal is actually larger than expected, it is perhaps important to consider objections raised by Chatfield and Harrison (1977). These authors claim that the earlier ozonesondes of the chemiluminescent type gave systematic lower ozone densities in the troposphere by about 50%. The majority of the above data have been published by Hering and Borden (1964, 1965a, b, 1967) and these data have been used in the Hilsenrath and Dunn (1979) as well as in the Mateer et al (1980) standard ozone profiles. For the  $\pm 30^{\circ}$  latitude range the chemiluminescent data represent almost 61% of the total number of soundings in the HD model. This point should be analysed in more detail.

The secondary ozone peak located in the troposphere is also significant from the theoretical point of view (Nicolet, 1975), since it represents the presence of a perhaps photochemical source of ozone in the troposphere. This possibility has been advocated by Fishman and Crutzen (1977) and Fishman et al (1979), suggesting as ozone sources in the troposphere the oxidation of CO, CH<sub>4</sub>, and other hydrocarbons in the presence of NO and NO<sub>2</sub>.

Figure 2a shows the height-time cross section of the data base. The curves are labeled in terms of ozone density in units of 10<sup>12</sup> molecules cm<sup>-3</sup>. The data base is still small to test, for example, for the biennial wave, but, from inspection of Figure 2a it appears that an increase in density in the layer as a whole is not coincident with the maximum peak density. To investigate this in some more detail we show in Figure 2b the correlation coefficient between the average yearly time series of the total abundance and the density at the peak, by time-shifting the position of the ozone maximum by respect to the total ozone abundance. It shows that the ozone total abundance lags behind the peak ozone density by about 90 days.

The yearly variations of total ozone content for the three years individually and the overall average are shown in Figure 3a. The data points have been smoothed by a six point running mean shown by the continuous lines whereas the actual data points are shown by crosses. On some occasions balloon disruption has occurred below the ozone peak and therefore the total number of ozone abundance

values is only 45 instead of 53. The dashed horizontal lines indicate the average of the data set.

A reasonable degree of recurrency can be seen from year to year. During the first 60 days of the year there are only six measurements. The ozone abundance at Natal seems to be decreasing during this period, reaching a minimum at about day 120. It should be noted, however, that the error bar derived at day 20 is rather large. The following increase seems to peak at a rather broad maximum centered at about day number 250.

It is interesting to note that the tropospheric ozone abundance, which represents only about 12% of the total abundance, is rather well correlated with the yearly variation of the total ozone content, giving a correlation coefficient of 0.62. This is probably due to higher tropospheric injection rates caused by higher densities in the region of the main  $O_3$  peak.

A comparison of the average ECC measured total content at Natal is made in Figure 3b (continuous line) with results from other measuring methods. An equatorial average of Nimbus 4 BUV data taken in the period 1970-71 (Hilsenrath et al, 1979) is shown as a dashed line. The average is only .248 cm STP. (The dashed line is upscaled by .030 cm STP units in the Figure). An average value close to .250 cm STP has been indicated also by Tolson (1981) from a longer series of the BUV data, and by Ramanathan (1963) from equatorial

Dobson data. The higher ozone contents at Natal are however confirmed by Dobson measurements.

The crosses show the average Dobson derived ozone total content at Natal (November 1978 to December 1981). It can be seen that the strong decrease during the first 60 days of the year indicated by the ECC data is not observed in the Dobson data. Because of the large scatter and scarcity of data points in the ECC data during this period more weight should be given, of course, to the Dobson data. The remaining features agree rather well. The Dobson overall yearly average is .275 cm STP and the sonde average is .287 cm STP.

The difference between the ozone profiles around the yearly minimum and maximum ozone total content is shown in Figure 3c. The minimum abundance (MIN) curve was computed from 13 measurement events that occurred between days 90 and 180, whereas the curve for maximum abundance (MAX) was derived from 16 measurement events between days 190 and 290. Typical mean square errors are shown as horizontal bars.

The increase in ozone abundance is due to an increase in density at all heights, and the overall increase in ozone is accompanied by a slight decrease of the peak height. This can be seen also in the larger 1980 densities of Figure 1a. It appears to indicate that larger total abundances are correlated with a lowering of the ozone peak, as already mentioned.

In Figure 3d the data is shown in different seasonal periods. As previously the horizontal bars represent mean square errors, in this case calculated for summer. In this season there are 16 measurements, whose average is shown by heavy dots. Summer has been defined as the months of November, December, January and February. For winter (May, June, July, and August) there are 17 measurements and the average profile is shown by open circles. For equinox (March, April, September, and October) there are 20 profiles and the average is shown by crosses. The total contents for the above periods are also shown in units of cm STP. They are respectively .293 for summer, .287 for winter, and .283 for equinox.

Figure 4 shows the variation of ozone and temperature, measured at the 100 mbar level, which is the average location of the tropopause at Natal. The actual values are plotted and a 4 point running mean (solid lines). A good degree of correlation can be seen. We have further investigated this relationship in the data, by calculating a correlation coefficient for the temperature and  $O_3$  yearly variations at several selected pressure levels. The result is shown in Figure 5. In the troposphere the correlation coefficient seems to be small but at the tropopause and up to the main peak of the layer the ozone is well correlated with the temperature. The correlation coefficient turns negative only above the height of the peak.

Below the ozone layer peak the positive correlation between ozone and temperature annual variations can be explained in

terms of vertical transport of air masses, since both ozone and temperature increase with height in this region. Above the peak, the negative correlation has been attributed to the inverse proportionality between the main photochemical ozone production (reaction rate) and temperature, but the high correlation found at the height of the peak (0.64 at 20 mbar) is perhaps unexpected. However, data from another station (Wallops Island) has also shown this positive high correlation slightly above the peak of the layer.

#### Summary

From the analysis of 53 balloon launchings, performed in a period of 3 years, in which ozone densities were measured through on board ECC sondes, the following points should be noticed:

1. Tropospheric ozone densities at Natal are higher by about 30% than previously published data in the equatorial region.
2. There seems to be a downward displacement of the ozone peak for larger total ozone contents. The displacement, however, is rather small.
3. The average ozone total content at Natal is higher by about 15% than the values quoted from measurements by satellites.

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### Figure Captions

Figure 1a. Yearly average profiles of ozone at Natal for 1979, 1980, and 1981. Number of balloon flights are shown in parenthesis. A biennial wave might be responsible for larger densities at the ozone peak in 1980.

Figure 1b. Average ozone profile based on 53 ECC soundings at Natal (continuous line). Total ozone content is .287 cm STP. The dash-dot line (HD) is an average ozone profile for .285 cm STP compiled at several stations situated within  $\pm 30^\circ$  latitude by Hilsenrath and Dunn (1979), shown for comparison.

Figure 2a. Height time cross-section for 53 ECC ozonesonde flights at Natal. Numbers on the curves represent ozone densities in units of  $10^{12}$  molecules  $\text{cm}^{-3}$ .

Figure 2b. Correlation coefficient between maximum ozone density and total ozone content. The phase shift for maximum correlation corresponds to about - 96 days and for minimum correlation 92 days.

Figure 3a. Yearly variations of ozone content. The crosses show the abundance values, the dashed lines show the average, and the continuous lines are 6 point running means.

Figure 3b. Comparison of the overall average Natal data repeated from previous figure (continuous line) to total ozone data determined by the Nimbus 4 BUW experiment during 1970-71 in the equatorial zone (dashed line) and to Dobson data taken also at Natal (crosses).

Figure 3c. Average profiles of ozone computed during the period of minimum total content (MIN) and during the period of maximum total content of ozone (MAX). Typical mean square errors ( $\pm \sigma$ ) are shown by horizontal bars.

Figure 3d. Data grouping for different seasonal periods. The ozone abundances in  $\mu\text{m STP}$  units are also shown.

Figure 4. Temperature and ozone variations at the 100 mbar level (tropopause). The dots are actual data points and the continuous smoother line is a 4 point running mean. The correlation coefficient is .45.

Figure 5. Correlation coefficient between temperature and ozone yearly variations at selected pressure levels, using 8 point running means for the T and  $\text{O}_3$  yearly data.

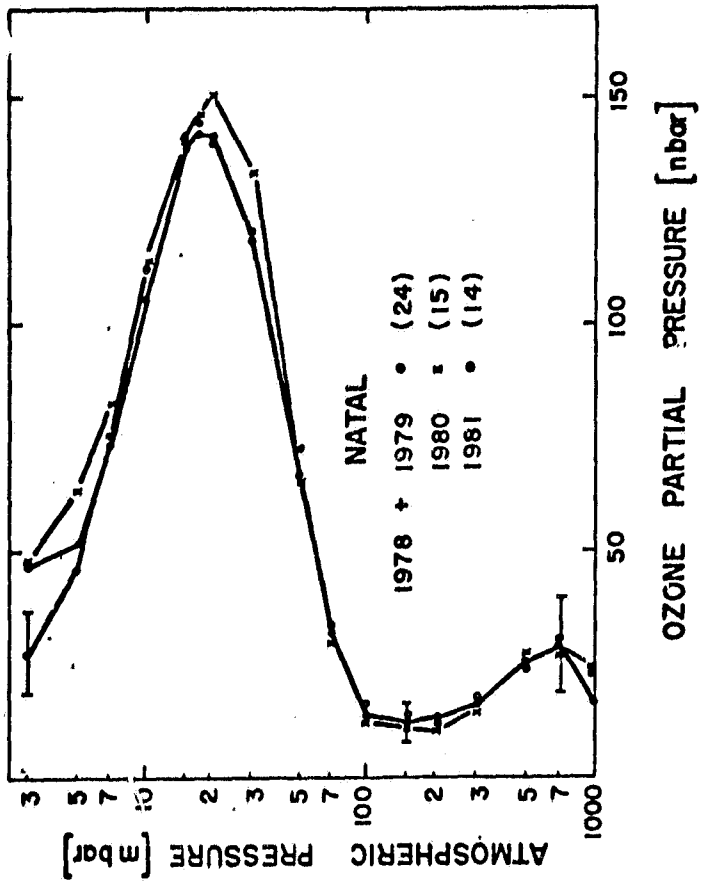


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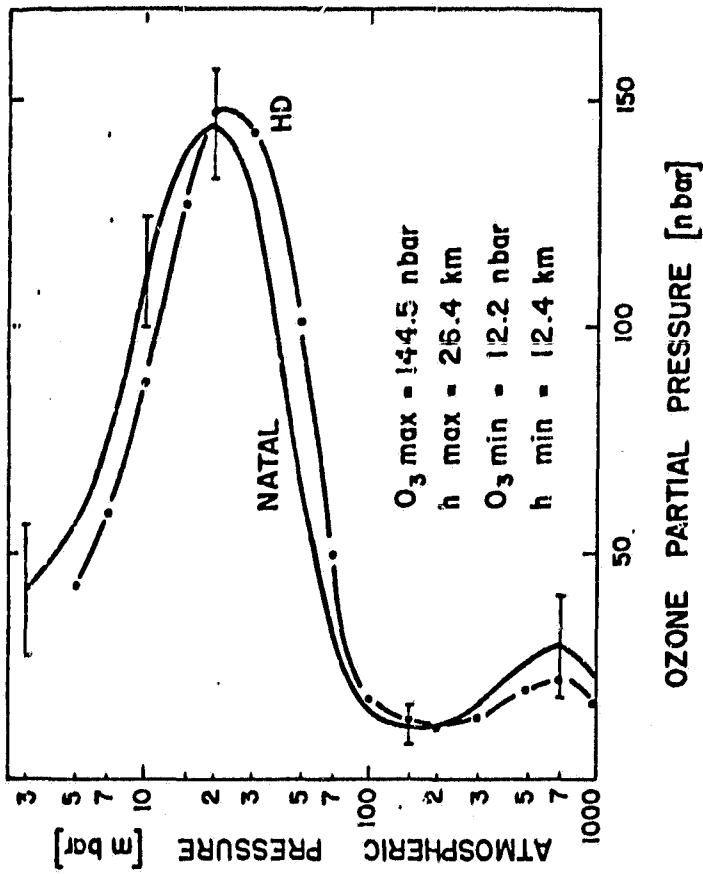


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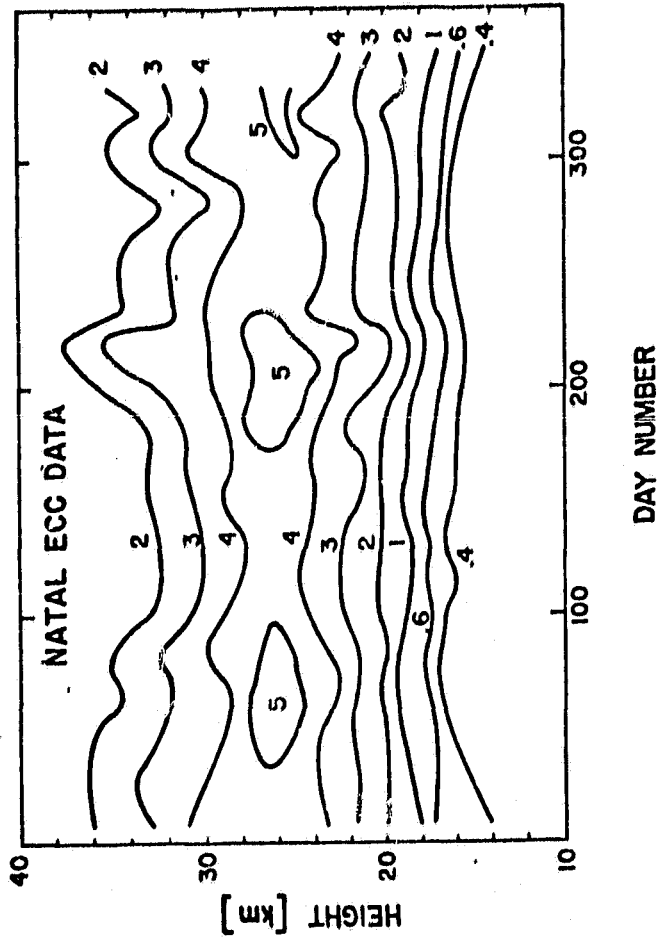


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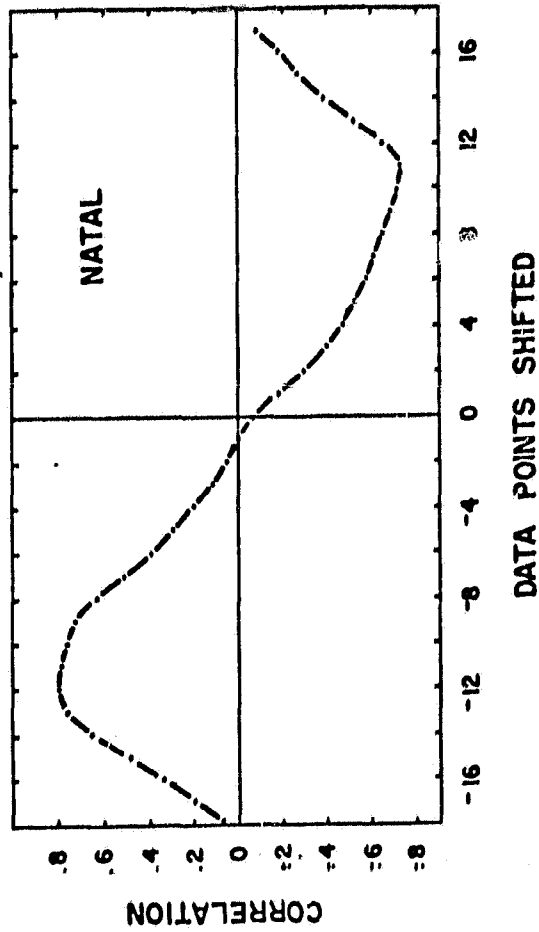


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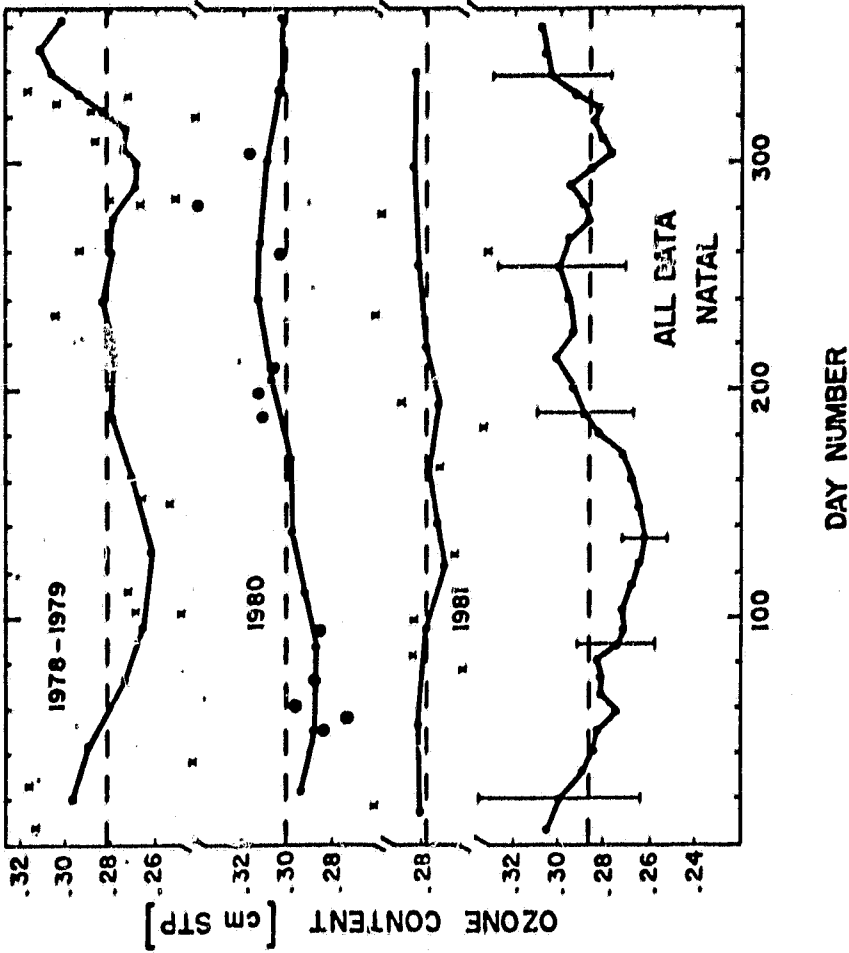


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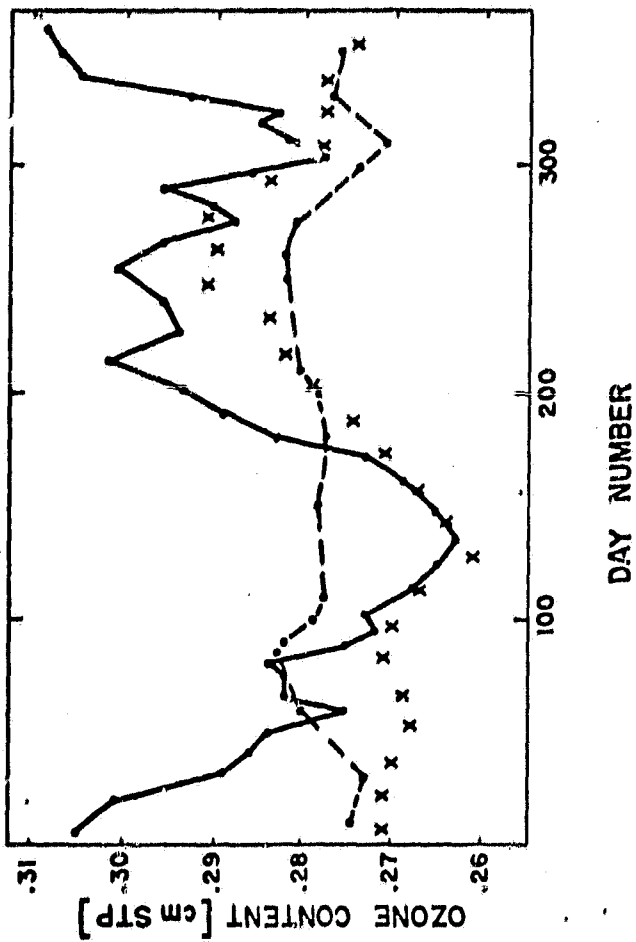


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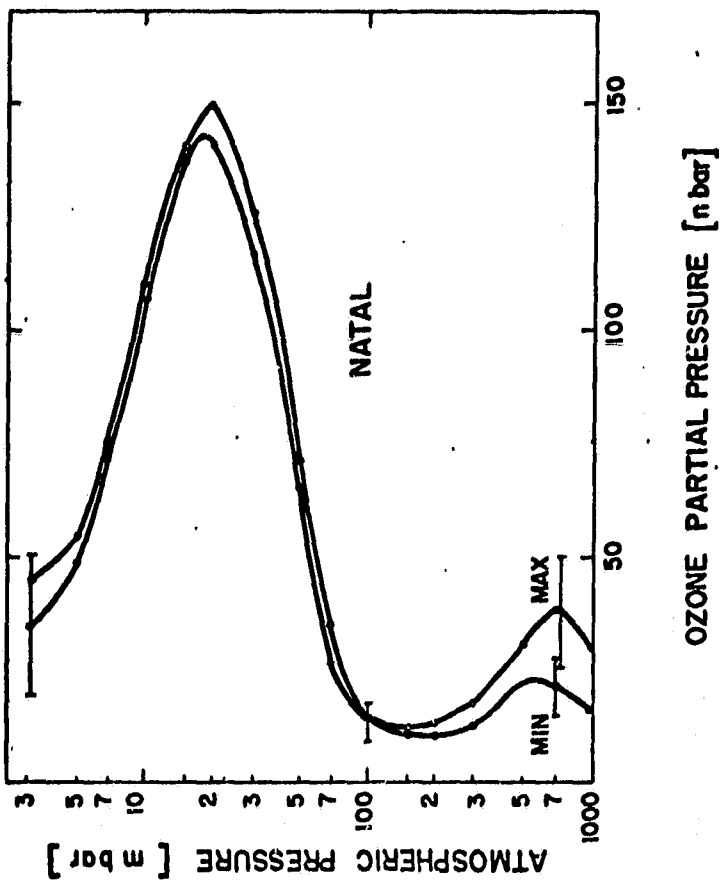


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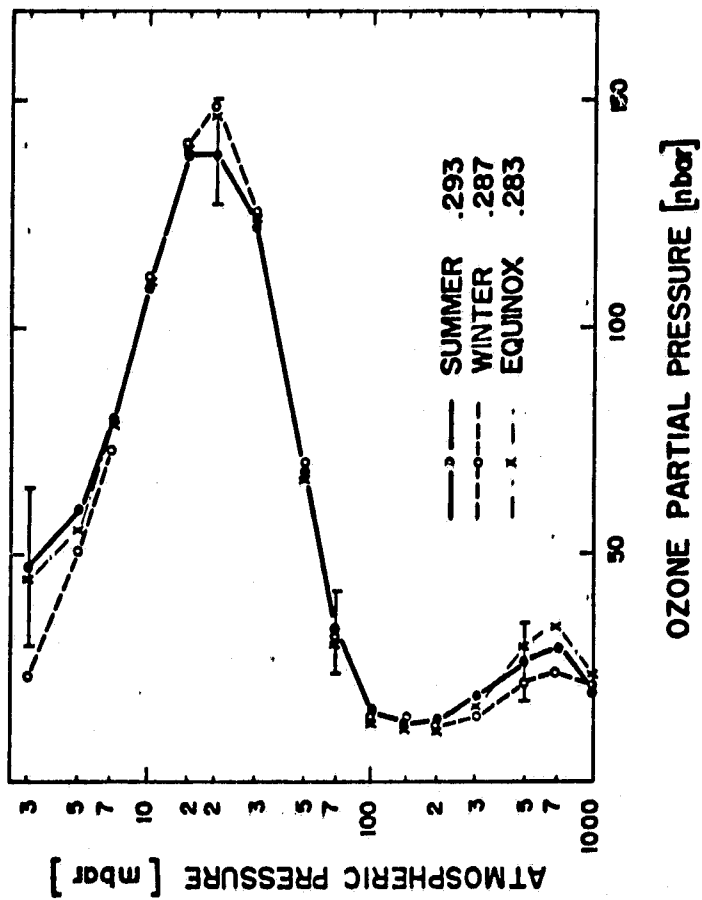


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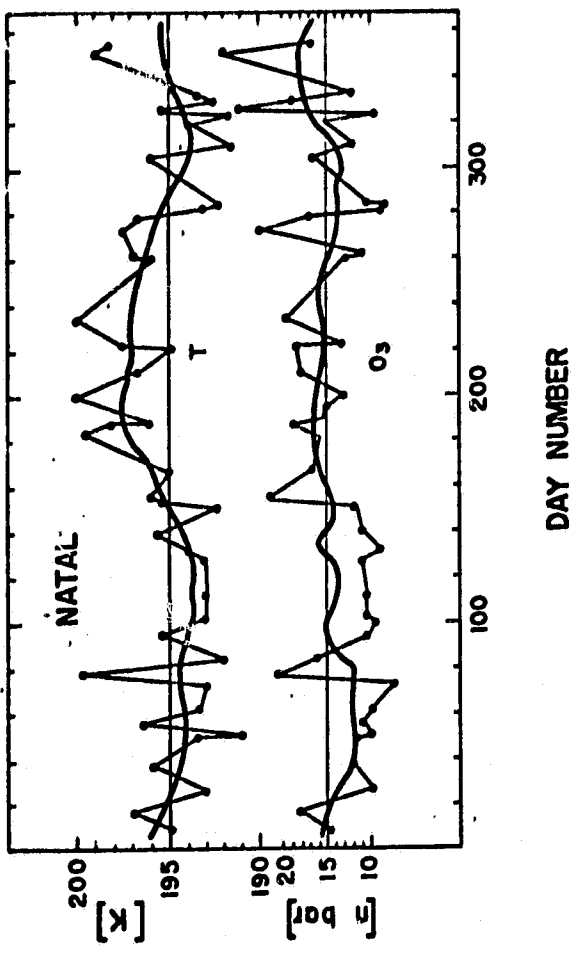


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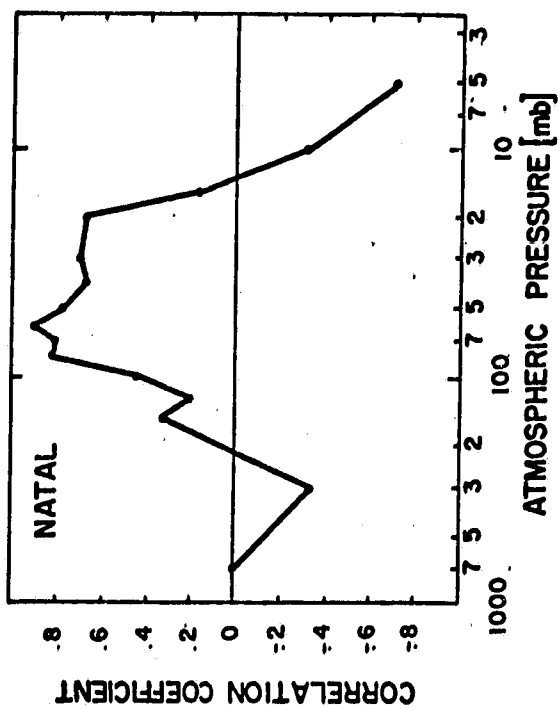


Figure 5. Correlation coefficient between temperature and ozone yearly variations at selected pressure levels, using 8 point running means for the T and O<sub>3</sub> yearly data.