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14. Abstract/Notes

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TROPICAL OZONE: SEASONAL VARIATIONS IN THE TROPOSPHERE AT 6° S

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ABSTRACT

Contrary to current wisdom a very strong seasonal variation of ozone concentrations is observed at Natal (6° S) with maxima in the months of September and October. Besides these large oscillations which can be seen in the whole troposphere, the average ozone conectrations of the lower troposphere are comparable in magnitude to those in Europe and parts of the USA, which is also unexpected on the basis of previous measurements in the tropics. These results have been obtained from 94 ozonesonde balloon releases.

May 1985

INTRODUCTION

This paper describes new features of ozonesonde data gathered at Natal (6° S, 35° W). These include a strong seasonal variation in the troposphere and a large ozone average concentration comparable to those found in the eastern U.S. and Europe. The seasonal variation in the troposphere is so large that it seems to drive the seasonal variation of the ozone total content, as seen for example by the Dobson spectrophotometer. These findings are not only new but they also contradict previous reports of small tropical averages and small seasonal variations.

Most of the known ozone data from the tropics were taken in the Panama canal zone (9° N). Previous studies are based on the data presented in Hering and Borden (1964, 1965 a, 1975 b, 1967).

The basic characteristic is that the number of soundings in the tropical region is strongly outnumbered by those at mid-latitudes. Only sparse measurements were made and also two different sondes were used with different sensitivities in the troposphere (Chatfield and Harrison, 1977 a), thus discouraging lengthier analyses. Nevertheless, Chatfield and Harrison (1977 b) used the available data to study seasonal dependencies along a meridional band, and Fishman et al. (1979) separated the data between Northern and Southern Hemispheres. The first authors found a seasonal variation for the Panama data with an average tropospheric mean that was low compared to the mid and higher latitudes, with seasonal peaks at the equinoxes. On the other hand, Fishman et al. (1979) conclude that tropospheric ozone in the tropics should be larger in the Northern Hemisphere.

The Natal data set is described in the next section. It has been taken in a consistent manner using the same technique, the same type of instrument, and the same observational group, and the total number of soundings now available is more than twice as large as for previous studies. The characteristics observed in the Natal data, as already mentioned, contradict the previous findings. Not only is the average ozone mixing ratio in the lower troposphere larger than that of the Northern Hemisphere tropical site, Panama (Kirchhoff, 1984), but it is even comparable to as high values as observed in North America and Europe, depending on season. Furthermore, the seasonal variation is very strong showing a maximum in spring and minimum in autumn, the maximum having almost twice as much ozone as the minimum.

It should be noted that recent instrument comparison campaigns (BOIC, Balloon Ozone Intercomparison Campaign, Ernest Hilsenrath, private communication) have shown evidence that presently used ozonesondes measure high ozone values in the troposphere when compared with UV photometers, by an amount of about 15%. The sondes used at Natal are calibrated at Wallops Island, where accuracy and precision tests gave the following results: (Arnold Torres, private communication) standard deviation averages in precision tests, less than 5 mb (15%), and accuracy less than 5 mb or 15%, in the troposphere. These tests include laboratory tests in a specially built environmental chamber (Barnes, 1982) and also dual sonde flights. In view of the large variations that we see in the Natal data, much larger than the above quoted values for accuracy and precision, we conclude that errors or biases of the sondes in the lower troposphere, as suggested by the BOIC campaign, will not affect the present data analysis.

DATA

Ozone partial pressures have been measured at Natal using the ECC ozonesonde (Komhyr and Harris, 1971). This instrument bubbles air through the chemical solution of the sensor and an eletric current is generated proportional to the ozone concentration. The ozonesonde is flown coupled to a standard radiosonde and thus almost simultaneous temperature, humidity, and wind data as a function of pressure can be obtained. At Natal a GMD station receives the 1680 MHz telemetry signal which is recorded on a strip chart. The measurements at Natal are the result of a programa of cooperation between the

National Aeronautics and Space Administration (NASA) of the U.S.A., and the Instituto de Pesquisas Espaciais (INPE) of Brazil.

A total of 94 balloon launchings has been considered in this study. Most of these measurements have been taken close to 1200 hours GMT. Part of this data has been described before in a preliminary report (Kirchhoff et al, 1981) and in a more general analysis (Kirchhoff et al, 1983) of results in the troposphere and stratosphere, using about one half of the present amount of data. Here we restrict our analysis to the troposphere since enough data is now available to examine the seasonal variation.

From 1978 to 1981 two launchings per month were generally scheduled. In 1982 only a few sporadic measurements were made, but since September 1981 four launchings per month have been made. The results from only three launchings have been rejected from this data analysis, because they showed unacceptably high ozone concentrations, beyond the average plus three standard deviations. Other seven profiles have not been considered because they were repetitions made on some days but at times other than close to 1200 hours.

RESULTS

Despite large typical day to day variations in tropospheric ozone a few common characteristics are apprent in the ozone profiles. Two different layers can be identified in the troposphere. The lower or planetary boundary layer which extends to about 600-500 mb, in which strong mixing may prevail, and a higher layer above, often called the free troposphere, which extends to about 200 mb. These layers can be easily identified in Figure 1, which is an average of data taken between 1978 and 1982. The horizontal bars are the standard deviation from the mean. The ozone mixing ratio increases with height in the boundary layer. It is perhaps important to note that most of the measurements having been made close to 0900 L.T. represent the conditions that prevail in a stable layer configuration. It is possible that an average, representing conditions close to local noon, may show a constant mixing ratio profile in the boundary layer, which would be expected for good mixing conditions. In the free troposphere, the ozone shows a roughly constant mixing ratio (see also Figure 2) but with considerable vertical structure in individual profiles. Large differences between fall and springtime can be seen in O_3 concentrations, humidity, and wind speeds, illustrated in Figure 2.

The average ozone mixing ratio profiles for the Natal data during spring and fall are shown in Figure 2, where the horizontal bars are again typical standard deviations about the mean. Data for the months 3, 4 and 5 were used to define the fall profile, and months 8, 9 and 10, for the spring profile. Also shown in Figure 2 are the average relative humidity profiles and the wind profiles. Most of the properties of these profiles have been mentioned earlier, but it is worth noting again that the middle troposphere has roughly a constant ozone mixing ratio, whereas the mixing ratio has a rather strong positive gradient above and below the free troposphere.

An additional important point to mention in Figure 2 is the comparison of the ozone profiles at Natal, a Southern Hemisphere tropical station, with those at mid latitudes in the Northern Hemisphere. Traditionally it has been assumed that ozone in the tropics is much less abundant than in mid latitudes, on the basis of earlier and very sparse data. In Figure 2, the dashed curves labeled F for fall and S for summer show the average ozone mixing ratio profiles at 38^{0} N. Clearly, depending on the time of year, the ozone profiles at 6^{0} S and 38^{0} N can be quite comparable. It should be also noted that at ground level, the mixing ratio of ozone varies between about 20 and 43 ppbv for data averaged from several European and U.S. stations (Logan, 1985; Logan et al, 1981).

At about 700-800 mb, the temperature often shows a constant or even positive gradient, an "inversion" and very close to this feature there is often a sharp peak in ozone and a simultaneous decrease in humidity, which may result from subsiding air. Higher convection activity in the wet season makes the boundary layer much higher reaching about 600-500 mb. The relative humidity (RH) has a much slower rate of decay with height, being 30% at about 300 mb. Over Natal, this occurs in autumn. The winds in this layer are predominantly westward, that is, from the ocean. The direction reverses towards the east at slightly above 500 mb in spring, and 400 mb in fall. During spring, individual profiles very often show a large positive gradient of the ozone mixing ratio just above the 800 mb pressure level which is coincident with strongly decreasing humidity and a simultaneous temperature "inversion". It appears that this is close to the upper limit of the boundary layer, below which strong mixing prevails, and on top of which masses of air are deposited by a large scale downward motion. The wind flow lines over Brazil are shown in Figure 3 for the 500 mb level. During spring Natal is very closely the center of a high pressure bulge of large scale anticyclonic wind motion (Sobral, 1980) which brings with it downward subsidence. This does not only dump mid-tropospheric air on the top of the boundary layer, but in doing so also has a tendency of drying the atmosphere. (Compare the spring and fall relative humidity profiles of Figure 2. The seasonal effect is also present here, of course. The rainy period occurs generally around February-March, and the dry period around August-September). Thus in the dryer periods the Natal area is characterized by subsidence,the atmosphere is practically dry above the upper part of the boundary layer, showing a very strong gradient of RH.

Above the boundary layer, in the middle troposphere, the ozone mixing ratio is roughly constant, but there is considerable structure, as already mentioned. We believe that this structure in mixing ratio is real and not data noise, on the basis that an average magnitude of the structure, being about 30% about the average, is much larger than either the accuracy or precision of the sonde quoted earlier. Further evidence is offered by profiles taken on subsequent days, which show the maintenance of parts of the structure for periods of the order of days.

A comparison of the tropospheric variations with those found in the stratosphere is shown in Figure 4. A very important characteristic is shown, namely that the variations in the stratosphere are very small compared to those of the troposphere. At 15 mb , for example, the relative variations of ozone about the annual average are less than ± 10% (uppermost curve in Figure 4) whereas in contrast there is a variation of about ± 50% around the yearly average at the lower levels.

It should be noted that this variability in the troposphere of Natal, where the values at the seasonal maximum are about twice as large as during the minimum, is much larger than that shown by data from Panama, a Northern Hemisphere tropical station. During spring, at 700 mb there is little difference and at 300 mb the spring values are abut 25% lower than during fall (Logan, 1985). The seasonal bahavior at Natal and Panama is therefore completely different.

The seasonal variation of the troposphere is further illustrated in Figure 5, where we show the integrated amount of monthly ozone averages. A simple average of all data has been used to normalize the seasonal variation. The variation observed with a Dobson spectrophotometer also installed at Natal, a few kilometers away from the balloon launching facility, is shown as well as the ozone integral of the troposphere, measured by the sondes between ground level and 100 mb. The continuous line is the Dobson result from a three year average of daily readings. It is normalized to 275 Dobson units. It is included in this Figure to show the seasonal variation of the total ozone integral in the troposphere and stratosphere. As shown, the maximum in September is slightly over 5% of the yearly average. This small seasonal variation shown by the Dobson data is typical for low latitude stations. The large dots are also measured by the Dobson instrument, and represent averages of those individual measurements made as close as possible to the ECC launchings. The normalization is the same as before. The other curves are integrals to only 100 mb measured by the ozonesondes, representing therefore only the troposphere. The variations are obviously much stronger. The open circles show straight averages. For the variations shown by X and squares special selections of the data have been made. In case of the X, the averages were calculated after all individual ECC profiles were normalized to the Dobson ozone integrals. The reason for doing this is not very clear but some workers feel that data that is not Dobson normalized is unreliable. The results are not substantially different from the straightforward averages except, perhaps, in April and August. Data points for May and December are missing but the strong seasonal variation in the troposphere is still evident. This is also true for the third case, shown by squares, in which we have eliminated data for which the ratio between the total content measured by ECC to the Dobson total content is outside the range 0.8-1.2. All three cases for the troposphere are normalized to 10^{18} molecules cm^{-2}.

With the results of Figure 5 one can argue that the density integrals of the troposphere, and their seasonal variations, drive the overall total ozone content (troposphere plus stratosphere) seasonal variation. Since most of the ozone is in the stratosphere the seasonal variation of the overall total content is normally ascribed to seasonal effects of the stratosphere. This strong seasonal variation of the total ozone content found at high and mid latitude decreases towards lower latitudes. Contrary to normal expectation, at Natal it seems that it is the tropospheric ozone integral which drives the overall total ozone content indicated by the Dobson instrument. The tropospheric variation is quite able to produce the 5% increase in total content shown by the Dobson data (continuous line). The overall total content average in molecules per cm^2 is 0.78×10^{19}. The amount of tropospheric ozone above the average is 0.03×10^{19} molecules cm^{-2} which makes up about the right size of the increase shown by the Dobson results (see Figure 5).

It has been suggested by Crutzen et al. (1979) that biomass burning may be a substantial source of atmospheric gases such as CO, and Seiler and Crutzen (1980) have further evaluated the influence of biomass burning on the atmosphere. Very large areas in the Brazilian cerrado and even deforested areas, following usual agricultural practice, are cleaned of the dead vegetation by burning it, especially in the dry periods. This could add substantial amounts of CO to the atmosphere above central Brazil. As pointed out, for example, in Fishman and Crutzen (1977), CO can act as precursor in the photochemical production of ozone. This relationship has been further investigated by Seiler and Fishman (1981).

The research mentioned above may have direct bearing on the Natal ozone measurements. As already mentioned the central Brazil area has an unusual wind circulation pattern (Figure 3). Can it transport the air from the source region, that is, the biomass burning areas of the cerrado, to the Northeast coast? The winds above 500 mb flow from the continent towards the coast and a large subsidence area there could be responsible for injecting the air downwards in the Natal region, an air enriched by several gases, produced many miles away over land.

Because of the relationship between CO and O_3 it would be of great value to start simultaneous measurements of CO and O_3. Planning for these extra activities is in progress.

CONCLUSIONS

From the analysis of 94 ozonesondes launched from Natal we conclude the following:

1) There is a strong seasonal variation of the ozone concentration in the whole troposphere of Natal. The maximum in September-October is about twice as large as concentrations in April-May.

2) The seasonal variation in the troposphere is so large that it is quite possible that it drives the overall ozone total content variation at this low latitude.

3) The average ozone concentration in the planetary boundary layer during spring is comparable to values found in Europe and the U.S.A.

4) The conclusions above are new and unexpected, on the basis of previous observations at other tropical stations. It is tempting to interpret these results in terms of an inland photochemical source with origin in biomass burning, brought over the Natal area by large scale circulation and subsidence characteristics as discussed in this paper.

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FIGURE CAPTIONS

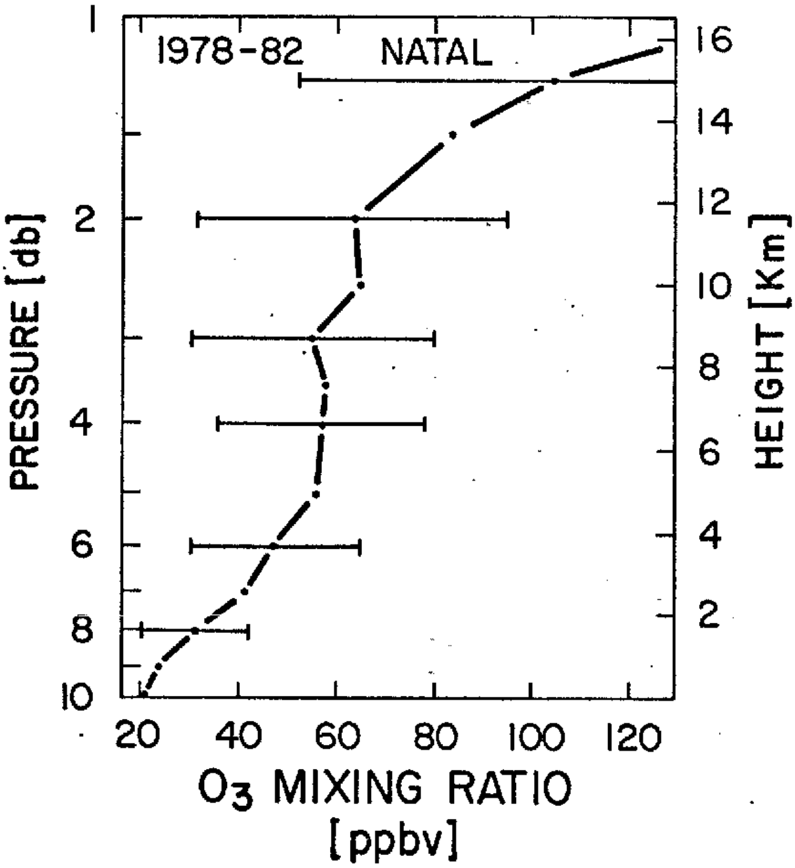
FIGURE 1 - Average mixing ratio profile in the troposphere of Natal. The concentration increases upwards in the planetary boundary layer, and is roughly constant in the free troposphere.

FIGURE 2 - Average mixing ratio profiles for spring and fall at Natal (heavy lines). Also shown are the average winds (dash-dotted) and humidity profiles (continuous lines) labeled F for fall and S for spring. For comparison, the dashed lines show average profiles for 38° N, with F for fall and S for summer.

FIGURE 3 - The wind flow stream lines at 500 mb over Brazil, showing a center of anticyclonic motion close to Natal giving origin to subsidence, during spring.

FIGURE 4 - Monthly average ozone concentrations at several pressure levels. In the lower levels of the troposphere a strong seasonal variation is present which disappears towards the higher altitudes.

FIGURE 5 - Monthly averages of ozone integrals in the troposphere (between ground level and 100 mb) and total content of ozone indicated by a Dobson photometer. The continuous heavy line shows a three year monthly Dobson average, and the dots indicate Dobson averages taken closest to the ECC launchings. The tropospheric integrals are straightforward averages shown by open circles, average tropospheric integrals after normalization to the Dobson total content (shown by X), and average tropospheric integrals after eliminating data outside the "correction factor" 0.8 and 1.2, shown by squares.



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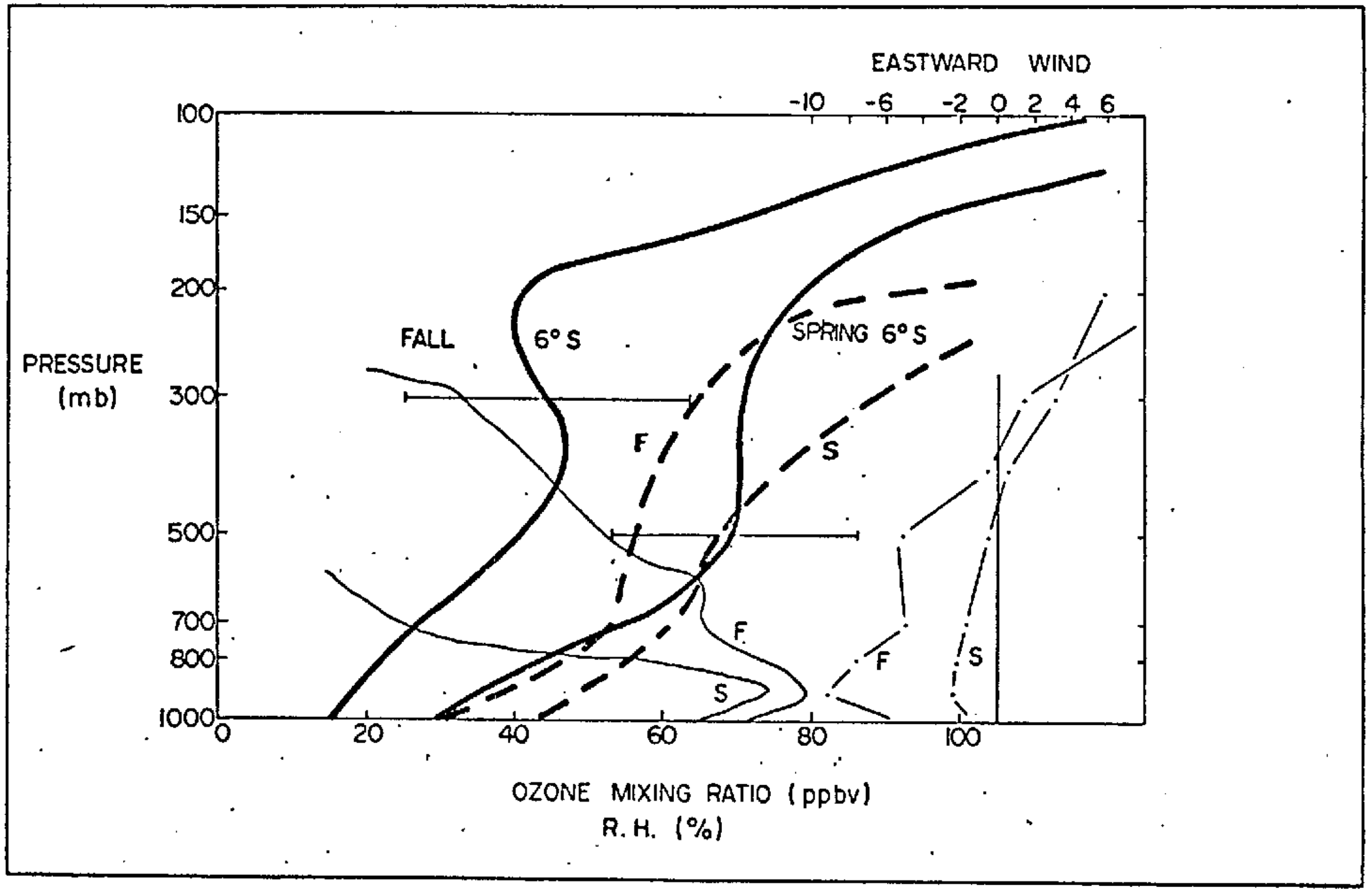

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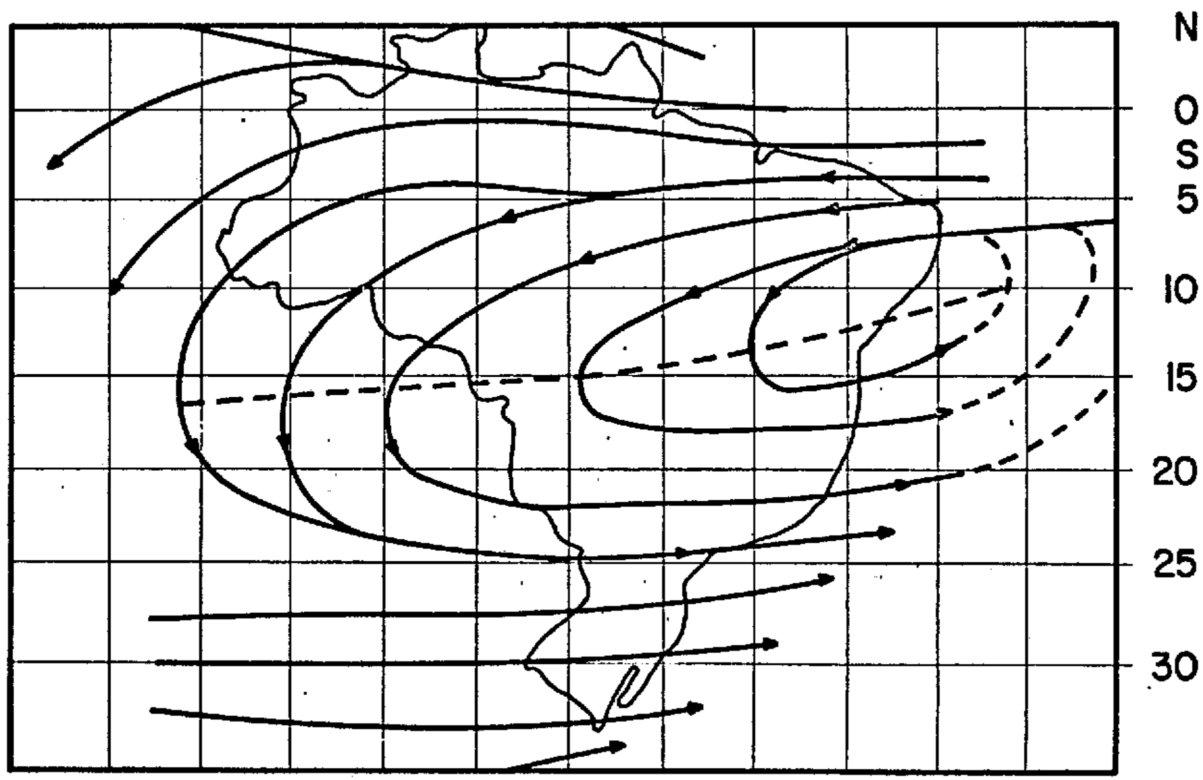

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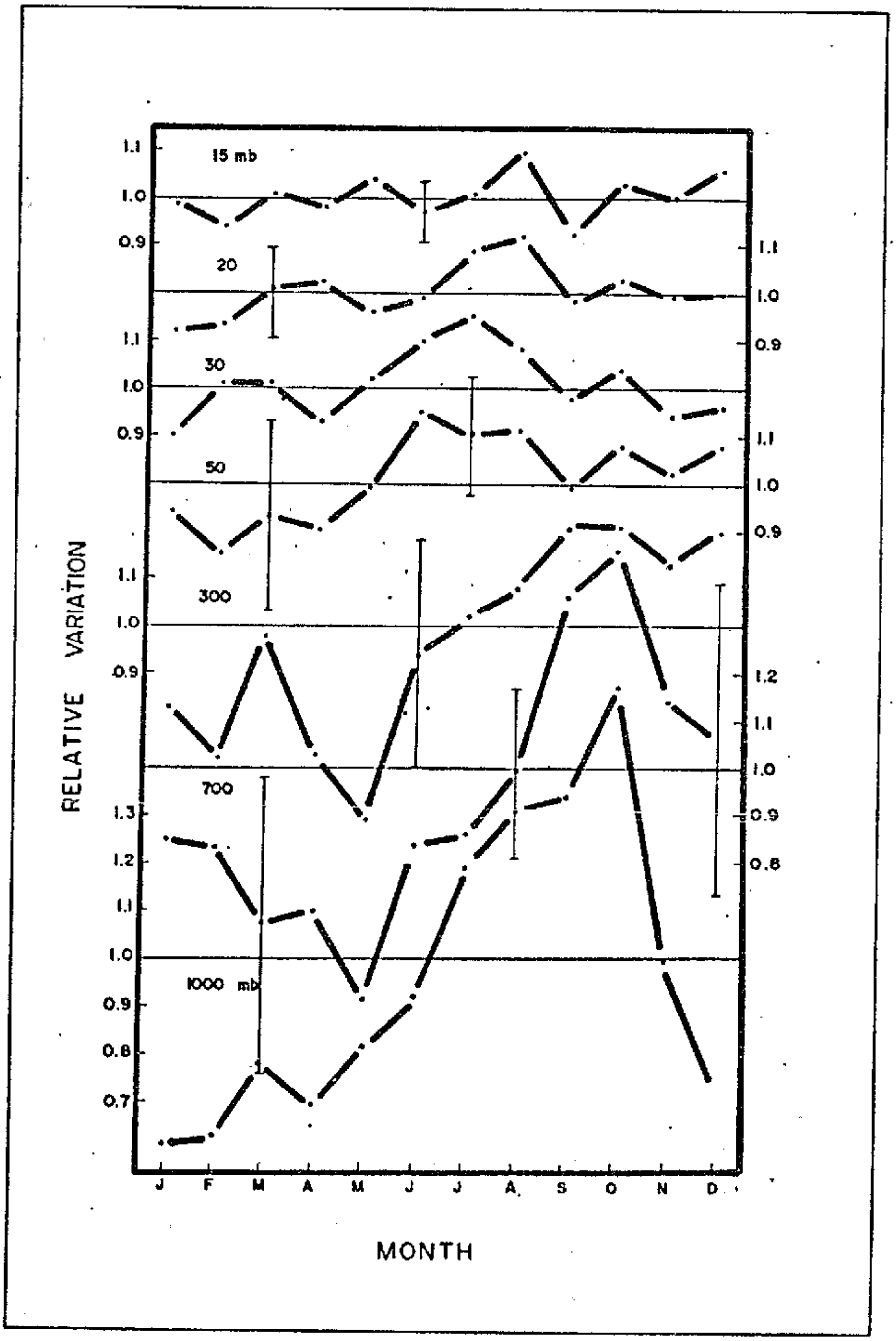

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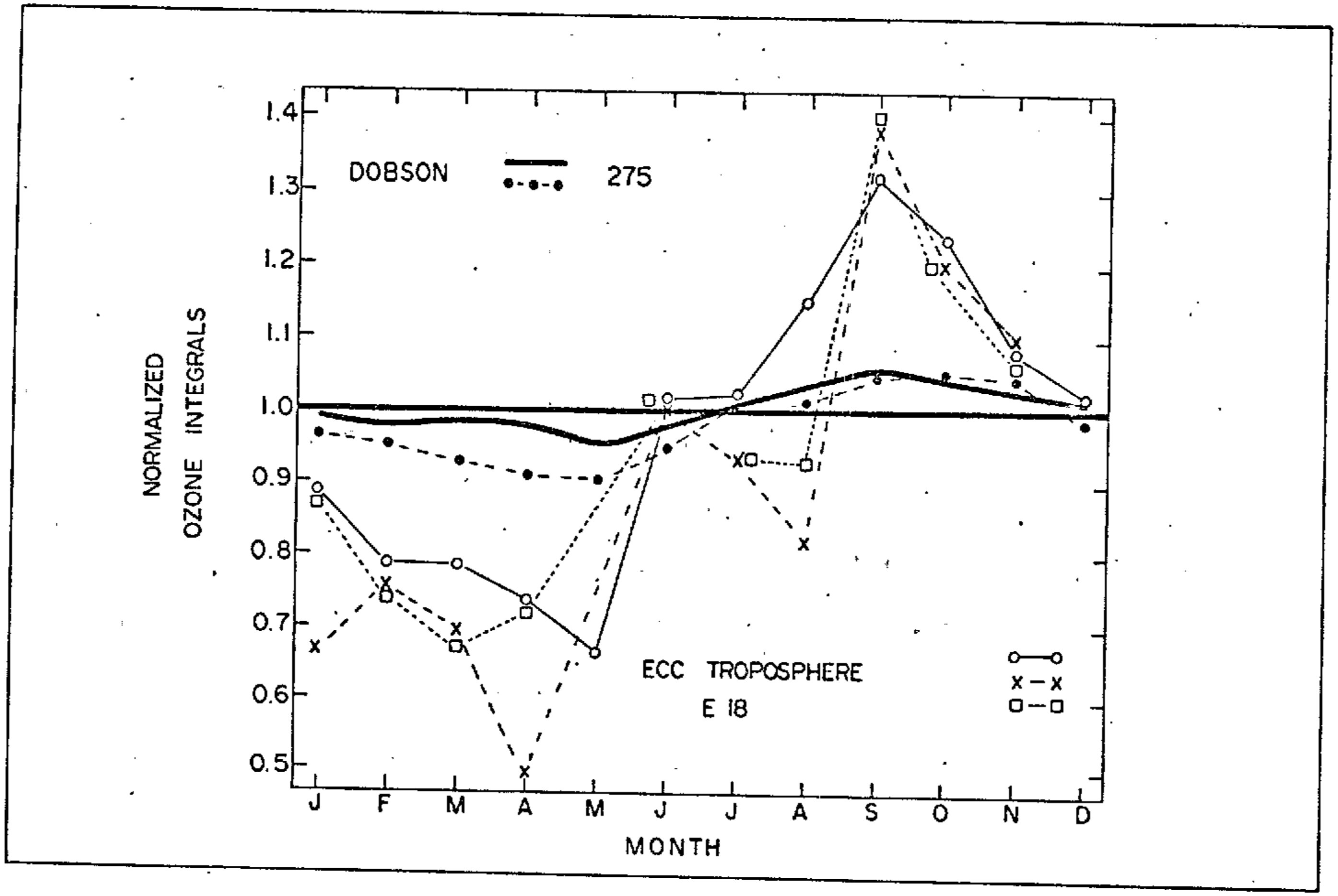

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REFERENCES

BARNES, R. The accuracy and precision of electrochemical concentration cell ozonesondes. Drexel University PH.D. Thesis, 1982.

CHATFIELD, R.; H. HARRISON. Tropospheric ozone, 1, evidence for higher background values. J. Geophys. Res., 82, 5965-5968, 1977a.

CHATFIELD, R.; H. HARRISON. Tropopheric ozone, 2, variations along a meridional band. J. Geophys. Res., 82, 5959-5976, 1977b.

CRUTZEN, P.J.; L.E. HEIDT; J.P. KRASNEC, W.H. POLLOK; W.SEILER.Biomass buring as a source of the atomospheric gases CO, H_2, N_2O, NO, CH_3Cl and COS, Nature, 282, 253-256, 1979.

FISHMAN, J.; S. SOLOMON; P.J. CRUTZEN. Observational and theoretical evidence in support of a significant in-situ photochemical source of tropospheric ozone. Tellus, 31, 432-446, 1979.

FISHMAN, J.; P.J. CRUTZEN. A numerical study of tropospheric photochemistry using a one dimensional model. J. Geophys. Res., 82, 5897-5906, 1977.

HERING, W.S.; T.R. BORDEN. Ozonesonde observations over North America. Vols 1-4, AFCRL-64-30, 2964, 1965a, 1965b, 1967.

LOGAN, J.A.; M.J. PRATHER; S.C. WOFSY; M.B. McELROY. Tropospheric chemistry: a global perspective. J. Geophys. Res., 88, 7210-7253, 1981.

LOGAN, J.A.. Ozone characteristics of the lower atmosphere, in preparation, 1985.

KIRCHHOFF, V.W.J.H. Are northern hemisphere tropospheric ozone densities larger? EOS 65, 449, 1984.

KIRCHHOFF, V.W.J.H.; Y.SAHAI; A.G. MOTTA. First ozone profiles measured with ECC sondes at Natal (5.9^{o}S, 35.2^{o}W) Geophys. Res., Lett., 8. 1171-1172, 1981.

KIRCHHOFF, V.W.J.H.; E. HILSENRATH; A.G. MOTTA; Y.SAHAI; R.A. MEDRANO B. Equatorial ozone characteristics as measured at Natal (5.9^{o}S, 35.2^{o}W). J-Geophys. Res., 88, 6812-6818, 1983.

KOMHYR, W.D.; T.B. HARRIS. Development of an ECC ozonesonde. Tech. Rep., ERL-APCL 18, Nat. Oceanic and Atmos. Admins., Boulder, CO, 1971.

SEILER, W.; P.J. CRUTZEN. Estimates of gross and net fluxes of carbon between the biosphere and the atmosphere from biomass burning. Climatic Change, 2, 207-248, 1980.

SEILER, W.; J. FISHMAN. The distribuition of carbon monoxide and ozone in the free troposphere. J. Geophys. Res., 86, 7255-7265, 1981.

SOBRAL, A.A. Um estudo climatológico dos campos de vento e de temperatura nos níveis superiores sobre a América do Sul. INPE-1672-TDL/017, 1980.