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#### Abstract:

The Transport and Atmospheric Chemistry Near the Equator-Atlantic (TRACE A) experiment. sponsored by the NASA GTE program, was a multinational field mission that took place simultaneously in Brazil, Africa, and the South Atlantic region, between the African and the Brazilian coasts. The general objective of the field mission was to investigate the tropospheric minor constituent composition, known to be disturbed by biomass burning practices. This report describes ozone measurements that were made by the Brazilian component. Two field missions in central Brazil were made with the objective of investigating ozone concentrations in the biomass burning source region: one smaller mission in the wet season period, April, and a major mission in the dry season, September/October The main field expedition during the dry season obtained data over a period of about 20 days in September and a few days in October 1992, in a savanna environment of central Brazil. Simultaneous surface ozone and ozone soundings were made. In the wet season the observation site was Goiania (16 degrees S, 49 degrees W); and in the dry season, two other sites were added: Cuiaba (16 degrees S, 56 degrees W) and Porto Nacional (11 degrees S, 48 degrees W). In addition, measurements were also made at an Atlantic coast site, Natal (6 degrees S, 35 degrees W), outside of the savanna region, and not affected directly by the biomass burning source areas, used as a control site. The average behavior of the ozone concentrations at the different sites suggests that surface ozone concentrations tend to be rather uniform, despite different precipitation rates, but slightly larger at the drier sites. However, other factors, such as burning fuel, for example, or cloudiness, may be also important to determine ozone concentrations. This is reflected by large day-to-day variabilities that are common in the source region. The diurnal variation of the surface ozone concentrations maximize around 1600 LT. Hourly averages in September, at this time, amount to 47 parts per billion by volume (ppbv) at Porto Nacional and 40 ppbv at Cuiaba. For this station the values are lower than those of previous years (55 ppbv in 1991 and 48 ppby in 1990). Only small differences, of the order of 5 ppbv, are observed between the source (burning) sites and Natal (the control site) in the wet season. In April, only 16 ppbv are observed at Natal. Much larger concentrations may be observed occasionally in the source areas, in the dry season. For example, at Porto Nacional, SO ppbv have been measured at the surface and in the lower troposphere. In comparison with the coastal site, near the surface, large scatter in concentration values at Porto Nacional (20-80 ppbv) contrast with the smaller concentration range seen at Natal (20-40 ppbv). In addition, at Natal the ozone mixing ratios below about 600 hPa are distributed around a vertical gradient in which the mixing ratios increase with height, whereas at Porto Nacional in the same height region, larger concentrations and a large scatter of the data are apparent. In the upper troposphere, perhaps surprisingly, the ozone concentrations at Natal and Porto Nacional are about equal, 70 ppbv at 10 km (with larger scatter at Natal than at Porto Nacional), probably reflecting a net production of ozone along

the pathways from the source regions, coupled with its longer lifetime at the higher altitudes. This data set is consistent with the hypothesis that tropical ozone in the troposphere is produced photochemically from biomass burning products in the dry season.

These products are exported from the source regions to the upper atmospheric levels by dry and wet convection and once in the upper atmospheric levels are taken eastward to the South Atlantic by the prevailing winds, where they contribute to local ozone formation. The air masses at Natal, Brazil, in the lower atmospheric levels, i.e., below about 500 hPa, originate from the South Atlantic. This allows one to classify Natal air masses as pristine over most of the year and justifies the stations use as a control station. However, the data now presented, combined with the detailed analyses of the other TRACE A studies, allows on to conclude that in the dry season, Natal ozone concentrations below about 500 hPa are perturbed by combustion products consistent with long-range transport from Africa.

#### **KeyWords Plus:**

TROPICAL SOUTH-ATLANTIC, TROPOSPHERIC OZONE, SURFACE OZONE, AMAZON BASIN, DRY SEASON, CITE-3, DISTRIBUTIONS, TRAJECTORIES, CLIMATOLOGY, ENVIRONMENT

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## Observations of ozone concentrations in the Brazilian cerrado during the TRACE A field expedition

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Abstract. The Transport and Atmospheric Chemistry Near the Equator-Atlantic (TRACE A) experiment, sponsored by the NASA GTE program, was a multinational field mission that took place simultaneously in Brazil, Africa, and the South Atlantic region, between the African and the Brazilian coasts. The general objective of the field mission was to investigate the tropospheric minor constituent composition, known to be disturbed by biomass burning practices. This report describes ozone measurements that were made by the Brazilian component. Two field missions in central Brazil were made with the objective of investigating ozone concentrations in the biomass burning source region: one smaller mission in the wet season period, April, and a major mission in the dry season, September/October 1992. The main field expedition during the dry season obtained data over a period of about 20 days in September and a few days in October 1992, in a savanna environment of central Brazil. Simultaneous surface ozone and ozone soundings were made. In the wet season the observation site was Goiânia (16°S, 49°W); and in the dry season, two other sites were added: Cuiabá (16°S, 56°W) and Porto Nacional (11°S, 48°W). In addition, measurements were also made at an Atlantic coast site, Natal (6°S, 35°W), outside of the savanna region, and not affected directly by the biomass burning source areas, used as a control site. The average behavior of the ozone concentrations at the different sites suggests that surface ozone concentrations tend to be rather uniform, despite different precipitation rates, but slightly larger at the drier sites. However, other factors, such as burning fuel, for example, or cloudiness, may be also important to determine ozone concentrations. This is reflected by large day-to-day variabilities that are common in the source region. The diurnal variation of the surface ozone concentrations maximize around 1600 LT. Hourly averages in September; at this time, amount to 47 parts per billion by volume (ppbv) at Porto Nacional and 40 ppbv at Cuiabá. For this station the values are lower than those of previous years (55 ppbv in 1991 and 48 ppbv in 1990). Only small differences, of the order of 5 ppbv, are observed between the source (burning) sites and Natal (the control site) in the wet season. In April, only 16 ppbv are observed at Natal. Much larger concentrations may be observed occasionally in the source areas, in the dry season. For example, at Porto Nacional, 80 ppbv have been measured at the surface and in the lower troposphere. In comparison with the coastal site, near the surface, large scatter in concentration values at Porto Nacional (20-80 ppbv) contrast with the smaller concentration range seen at Natal (20-40 pphy). In addition, at Natal the ozone mixing ratios below about 600 hPa are distributed around a vertical gradient in which the mixing ratios increase with height, whereas at Porto Nacional in the same height region, larger concentrations and a large scatter of the data are apparent. In the upper troposphere, perhaps surprisingly, the ozone concentrations at Natal and Porto Nacional are about equal, 70 ppbv at 10 km (with larger scatter at Natal than at Porto Nacional), probably reflecting a net production of ozone along the pathways from the source regions, coupled with its longer lifetime at the higher altitudes. This data set is consistent with the hypothesis that tropical ozone in the troposphere is produced photochemically from biomass burning products in the dry season. These products are exported from the source regions to the upper atmospheric levels by dry and wet convection and once in the upper atmospheric levels are taken eastward to the South Atlantic by the prevailing winds, where they contribute to local ozone formation. The air masses at Natal, Brazil, in the lower atmospheric levels, i.e., below about 500 hPa, originate from the South Atlantic. This

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allows one to classify Natal air masses as pristine over most of the year and justifies the stations use as a control station. However, the data now presented, combined with the detailed analyses of the other TRACE A studies, allows one to conclude that in the dry season, Natal ozone concentrations below about 500 hPa are perturbed by combustion products consistent with long-range transport from Africa.

#### Introduction

#### **Biomass Burning**

Large biomass burning events are common in central Brazil, the so-called Brazilian "campo cerrado" (cerrado, for short notation) region, a savanna type environment, with grasslands, shrubs, and small trees, of which large areas are set on fire every year. It is interesting to comment that fires in these areas, contrary to those used to clear forest areas, do not contribute to the greenhouse effect, since most of the dry vegetation set after will regrow the next year. These burnings, however, do contribute strongly to modify the natural composition of the lower atmosphere, by increasing the concentration of several gases, one of the most important of which is ozone (O3) [Kirchhoff et al., 1989; Kirchhoff and Marinho, 1994]. In a companion paper, Kirchhoff and Alvalá [this issue] describe other characteristics of deforestation and biomass burning, and it is shown that the burning pattern in Brazil has changed in recent years, in intensity and in geographic location.

#### TRACE A Project

First discussions on the Transport and Atmospheric Chemistry Near the Equator-Atlantic (TRACE A) were started in 1987, when it was felt desirable to make a comprehensive field expedition to study the global impact of gases emitted by large biomass fires in the tropics. Further interest was added when a large ozone maximum detected by satellite sensors (TOMS and SAGE [Fishman et al., 1990, 1991]) had been observed over a very large area in the South Atlantic region, between the South American and the African continents. This ozone bulge, an excess ozone amount in the troposphere, became one of the prime foci of the TRACE A mission. Because of the global dimensions of the ozone maximum it became clear from the beginning that an aircraft component for the mission would be essential, and eventually the NASA DC-8 was used for the oceanic segment. The TRACE A experiment is described in the overview paper by Fishman et al. [this issue].

Besides the South Atlantic region, where the tropospheric ozone maximum was detected, two other geographic areas of great interest to the project were the biomass burning areas of Brazil and Africa. These seem to have a direct bearing on the ozone maximum of the South Atlantic, namely, they are thought to be source regions, where the primary precursor gases are produced through biomass burning, which are then exported to the South Atlantic region by air mass transport. This hypothesis was first analyzed for Natal ozone data lKirchhoff and Nobre, 1986; Logan and Kirchhoff, 1986; the ozone seasonal variation at Natal is compatible with a distant (and thus indirect) biomass burning driving source. The same origin has been attributed to the South Atlantic ozone bulge of Fishman and coworkers [Fishman et al., 1990, 1991]. In addition to the biomass burning hypothesis the DC-8 mission also investigated a possible contribution from a stratospheric source. However, such a high-level ozone source was only found outside of the tropic belt, around 30°S, and is described by Browell et al. [this issue]. In any case, if biomass burning is the primary ozone source, it would also be important to know details on the location and distribution of the sources, and therefore the field expeditions in Brazil and Africa became additional segments of the TRACE A mission.

#### Measurement Background

Previous work investigating the result of biomass burning on the lower atmosphere has shown that CO and O3 increase their concentrations in the troposphere in the burning season, as compared to their values in the wet season [Crutzen et al., 1979, 1985; Delany et al., 1985]. These pioneering findings were based on in situ research campaigns using aircraft. They stimulated the formation of new atmospheric chemistry groups in different parts of the world and made possible further investigations in this area of research. To cite a few examples in the tropics, local smaller-scale field campaigns in Amazonia followed, such as the Cerrado 90 and Tocantins 91 field missions [Kirchhoff and Marinho, 1994] and the large international field missions CITE 3 [Anderson et al., 1993; Hoell et al., 1993; Andreae et al., 1994], ABLE 2A and 2B [Gregory et al., 1988; Kirchhoff, 1988; Kirchhoff et al., 1988, 1990; Browell et al., 1988. 1990], as well as the more recent TRACE A mission, in Brazil [Kirchhoff et al., 1992b], in the South Atlantic, and in Africa (SAFARI), and the previous DECAFE mission in Africa | Andreae et al., 1992]. Not just sporadic field missions but also long-term ground-based monitoring work was started, for example, in Brazil [Kirchhoff and Rasmussen, 1990; Kirchhoff et al., 1989], in Africa [Cros et al., 1987, 1988], in Venezuela [Sanhueza et al., 1985], and in tropical Asia [Ogawa and Komala, 1988]; and a collection of shipboard data is available from over the Atlantic Ocean [Winkler, 1988].

The situation was quite different with respect to ozone soundings in the tropics. It appears that only Natal, Brazil, has maintained systematic and continuous soundings in the tropics since 1978 [Kirchhoff et al., 1991], with a large campaign in the Amazonian forest during the ABLE missions, and sporadic measurements in the savanna region. Important sonde work was also started in Africa in preparation for and during the TRACE A mission [Cros et al., 1992; Olson et al., this issue]. The results reported in this paper are therefore an important contribution describing tropospheric ozone in the cerrado region of Central Brazil, during an intensive field campaign in 1992.

#### Objective of Experiment

The main objective of the field work in Brazil was to characterize the biomass burning source area in terms of the concentration of ozone. The major questions addressing the ozone concentration levels in the biomass burning source regions are discussed in the Results section. One of the early concerns was to better identify the source area itself by answering the following question: Where are the major source regions located in Brazil? It is important to realize that the pattern of biomass burning in Brazil has changed in the past few years. From analysis of the National Oceanic and Atmospheric Administration advanced very high resolution radiometer (NOAA)

AVHRR) data a large number of fires in the dry season used to be located in Rondonia, a tropical rain forest environment in the west of Brazil, where a number of large agricultural developments took place. This was partly the result of incentives from the Federal Government, which meant to develop Amazonia. This activity has slowed down considerably, in Rondonia and elsewhere, and presently the fire frequency maximum has shifted to the cerrado region of the state of Tocantins, especially the northern part. In addition, the deforestation rates, according to an INPE 1992 study, have been declining from rates of 21,130 km<sup>2</sup>/yr for the period 1978-1989, to rates of 17,860 for 1988-1989, 13,810 for 1989-1990, and 11,130 for 1990-1991, respectively, and it is estimated that present rates (for 1995) are somewhat below 10,000 km<sup>2</sup>/yr. Kirchhoff and Alvalá [this issue] show the major contribution of the number of fires, as detected by NOAA AVHRR, by state, in 1992. The largest percentage of fire foci, 22%, belong to the state of Tocantins. Second in burning was the state of Mato Grosso, with 19%. The choice of measurement sites for TRACE A Brazil was partly influenced by this characteristic.

The total number of fire counts, for the period between 1990 and 1993, shows three major and more consistent cluster points for biomass burning in Brazil: in the north of the state of Mato Grosso, around (11°S, 55°W), near the border with the state of Pará, where extensive yearly burnings occur with a considerable proportion of slashed forest burning, producing smoldering combustion; in the north of the state of Tocantins, around (8°S, 45°W), in a region of cerrado; and, finally, near the border of the states of Tocantins and Pará, around (5°S, 50°W), where again part of the burnings is slashed forest.

Additional specific questions addressed in this study are as follows: (1) What are the  $O_3$  concentrations in the source regions as compared to those observed outside? Here, the Natal station is used as an "outside" monitoring site. (2) Are there differences in concentration among biomass burning sites

Table 1. Cuiabá (16°S, 56°W) 1992 Ozonesonde Launches for TRACE A Brazil

Number	Date	Universal Time (Hours, Minutes)
l	Sept. 18	1936
2	Sept. 19	1530
2 3	Sept. 20	1314
4	Sept. 21	1504
5	Sept. 22	1834
6	Sept. 23	1433
7	Sept. 24	1654
8	Sept. 25	1700
9	Sept. 26	1700
10	Sept. 27	1713
<b>H</b> 1	Sept. 28	1708
12	Sept. 29	1705
13	Sept. 30	1657
14	Oct. 1	1651
15	Oct. 2	1658
16	Oct. 3	1657
17	Oct. 4	1604
18	Oct. 5	1629
19	Oct. 6	1856
20	Oct. 7	1705
21	Oct. 10	1412
22	Oct. 10	1618

All VIZ/ECC digital sondes.

Table 2. Porto Nacional (11°S, 48°W) 1992 Ozonesonde Launches for TRACE A Brazil

Number	Date	Universal Time (Hours, Minutes)
ı	Sept. 15	2053
2	Sept. 16	1151
3	Sept. 17	1949
4	Sept. 19	1642
5	Sept. 21	1638
6	Sept. 22	1400
7	Sept. 23	1209
8	Sept. 23	1810
9	Sept. 24	1830
10	Sept. 25	1752
11	Sept. 26	1730
12	Sept. 27	1838
13	Sept. 28	1404
14	Sept. 29	1336
15	Sept. 30	1404
16	Oct. I	1752

All Vaisala/ECC digital sondes.

in the same seasonal period? How heterogeneous or homogeneous is the region in terms of the ozone concentration in the same seasonal period? (3) Does upper tropospheric ozone behave seasonally as surface ozone? Given the different time constants and production rates involved, how are the variations at different height levels? (4) Are the 1992 observations similar to previous years? How does TRACE A results compare with previous years? (5) How do the source region O<sub>3</sub> profiles compare to the Natal climatology in the wet and dry seasons?

#### Experiment

#### **Activities in Brazil**

The TRACE A Brazil experiment had two components in terms of timing: a dry season mission and a wet season campaign. During the major dry season mission, in September, 72 ozone soundings were made: 22 at Cuiabá, specified in Table 1; 16 at Porto Nacional, shown in Table 2; and 34 at Natal, displayed in Table 3. For the wet season mission of April 1992 the Goiânia site was used for making eight ozone soundings in addition to surface observations, as shown in Table 4.

Ozone soundings during TRACE A were also made at Ascension Island and in Africa and are described in detail by Olson et al. [this issue]. This report describes part of the measurements that were made in Brazil, in the burning source area and near the Atlantic coast. A companion paper describes an aircraft mission, with the comparison of wet and dry season results of several trace gases, in 1992 [Kirchhoff and Alvalá, this issue]. The objective of this paper is to describe the tropospheric ozone observations.

Major TRACE A activities in Brazil were daily radio soundings at 16 stations; a special additional radiosonde network of six stations that made a special campaign of three soundings a day for studies of local convection; observations of ozone, carbon monoxide, methane, carbon dioxide, and nitrous oxide, and black carbon onboard two Bandeirante aircraft; observations at three surface stations of the parameters mentioned above; and ozone soundings, in the troposphere and stratosphere, at three stations.

Table 3. Natal (6°S, 35°W) Ozonesonde Launches in 1992 From 12 to 25 Vaisala/ECC Digital Sondes

Number	Date	Universal Time (Hours, Minutes)
1	Jan. 3	1600
2	Jan. 13	1554
3	Feb. 7	1600
2 3 4 5 6	Feb. 17	1640
5	Feb. 27	1414
6	March 5	1228
7 8	March 14	1600
8	March 25	1602
9	April 3	1330
10	May 8	1300
11	May 19	1315
12	Aug. 17	1133
13	Aug. 17	1539
14	Aug. 24	1948
15	Sept. 9	1245
16	Sept. 15	1407
17	Sept. 21	1432
18	Sept. 23	1340
19	Sept. 24	1327
20	Sept. 27	1627
21	Sept. 28	1429
22	Sept. 28	1701
23	Sept. 28	1945
24	Sept. 28	2120
25	Sept. 30	2003
26	Oct. 2	1330
27	Oct. 9	1334
28	Oct. 15	1905
29	Oct. 18	1902
30	Oct. 21	1830
31	Oct. 22	1930
32	Oct. 23	1735
33	Nov. 11	1636
34	Dec. 23	1632

Remaining sondes are VIZ/ECC analog sondes.

#### **Experiment Sites**

The measurement sites used for TRACE A Brazil are shown in Figure 1. The major sites for ozone measurements are designated by squares. The circles show the location of sites used for radio soundings. Figure 1 shows three sites in central Brazil: Cuiabá (16°S, 56°W), Goiánia (16°S, 48°W), and Porto Nacional (11°S, 48°W). All sites are located in a cerrado environment, but Cuiabá has the influence of the rain forest area to the north and west. The INPE group has made ground observations of O<sub>3</sub> and CO at this station for several years [Kirchhoff et al., 1989, 1992a; Kirchhoff and Rasmussen, 1990]. The other

Table 4. Goiânia (16°S, 49°W) Ozonesonde Launches in April 1992 (Wet Season)

Number	Date	Universal Time (Hours, Minutes)
1	April 8	1515
2	April 9	1417
3	April 10	14 t 3
4	April 11	1324
5	April 12	1315
6	April 13	1426
7	April 14	1343
8	April 15	1335

Vaisala/ECC digital sondes.

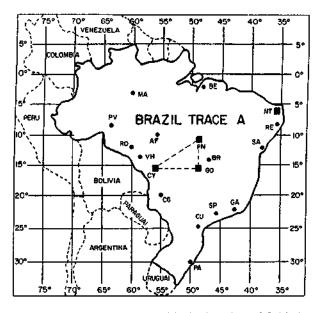


Figure 1. The map of Brazil with the location of field sites used during TRACE A. The squares denote sites for ozone work, the circles show radiosonde sites.

two stations, Goiânia in the state of Goiás and Porto Nacional in the state of Tocantins, are relatively new observation sites. Goiânia is closer to a number of agricultural developments, with large soybean plantations, for example, whereas Porto Nacional is a more remote location, where the largest activity is raising cattle.

A fourth important data-collecting site in Brazil is Natal (6°S, 35°W). This site has been used routinely for ozone soundings since 1978 [Kirchhoff et al., 1991]. It is a constal site which receives a continuous strong sea breeze, which makes it useful as a control site for biomass burning studies [Kirchhoff et al., 1992a]. Special soundings were made at Natal for the TRACE A mission (as well as at Ascension Island and in Africa [Olson et al., this issue]), and special radio soundings were made for the study of vertical convection and redistribution of source gases [Pickering et al., this issue].

#### Instruments and Methods

Electrochemical concentration cell (ECC) ozonesondes have been used to measure ozone in the troposphere and stratosphere. The ozonesonde is a small balloon-borne device developed at NOAA [Komhyr, 1969; Komhyr and Harris, 1971]. The sensor itself is a small cell with two platinum electrodes immersed in iodide solutions of different concentrations in the cathode and anode chambers. When air containing ozone is circulated through the cathode, an electric current is generated which is telemetered to the ground receiver. The ozonesonde is flown in parallel with a standard radiosonde, which provides measurements of ambient air pressure, temperature, instrument temperature, and humidity. Winds may also be obtained through either Omega, Loran, or radar-tracking systems. The technique to prepare and launch these ozone sensors has been used regularly in Brazil since 1978, in a NASA-INPE long-term collaboration program [Kirchhoff et al., 1991]. The ozonesondes that were used in this experiment were calibrated in the laboratory with UV ozone photometers, whose calibrations were traceable to the photometric ozone standard of the National Institute of Standards and Technology. The accuracy

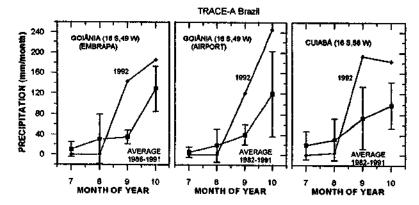


Figure 2. Precipitation at Goiânia and Cuiabá, showing that considerable more precipitation was seen in September and October 1992 than one would expect on the basis of the climatological averages.

and precision of different kinds of ozonesondes have been determined, for example, by Barnes et al. [1985] and Hilsenrath et al. [1986]. The following ground-based equipment was used: a DACOS digital system from Vaisala, at Porto Nacional, and Natal; a W-9000 digital system from VIZ at Cuiabá; and a SARA system for 403-MHz analog sondes, used at Natal. For the in situ measurements of ozone near the surface, ozone UV absorption photometers were used, calibrated as above.

#### Atmospheric Conditions during TRACE A

The TRACE A mission in Brazil, the South Atlantic, and Africa was planned for the peak dry season period. However, mechanical trouble with the NASA DC-8 delayed the start of the mission by 15 days. This advanced the mission toward the end of the dry season. It was found that the two sites Cuiabá and Golânia, had much higher precipitation rates in September and October 1992 than the climatological mean. This is shown in Figure 2. For Goiânia, two rain gauge stations were available, one at Embrapa and one at the local airport. The previous record, from 1986 to 1991 at Embrapa, defines the average behavior at this station. For the local airport the available data record spans a period from 1982 to 1991. The error bars are the standard deviations from the mean and reflect the year-to-year variability at each station. The data for Goiânia clearly show that in September 1992 the precipitation intensity was 3 times as large as the "climatological" average. For October the precipitation amount was also considerably larger than the average. The third panel in Figure 2 shows precipitation results for Cuiabá. Here again the precipitation in September and October 1992 was at least a factor of 2 larger than the climatological average. Evidently, this result of high precipitation rates in September and October 1992 at Cuiabá and Goiânia affected the overall results in terms of biomass burning intensity in the region, as will be shown again below examining the amount of fires detected by satellite. It is also interesting to note that contrary to September and October the months of July and August were effectively very dry months at Cuiabá and Goiânia.

The third station used in this study was Porto Nacional. Contrary to what occurred at Cuiabá and Goiânia this site was quite dry during September 1992. The strong cold fronts that were able to reach Cuiabá and Goiânia dissipated before they could propagate as far north as Porto Nacional. The precipitation rate for September, at Porto Nacional, amounted to slightly less than one half of the "climatological" mean, defined

with an 11-year data sequence available from 1977 to 1987. This is detailed in Figure 3, which shows a typical complete seasonal pattern of dry and wet seasons. The region is extremely dry during 3 months of the year, June, July, and August, with less than about 10 mm/month. May and September are also still very dry, with less than 50 mm/month of rain. Notice also that the transition time between wet (300 mm/month) and dry periods is rather short, only about 1 month.

In general, Meteosat images show an almost cloudless panorama over central Brazil during the dry August-September period in the Brazilian cerrado. This was also the case for 1992, during August, but in September, cold fronts advanced from the southwest as far as Cuiabá and Goiânia but not reaching Porto Nacional.

Figure 4 shows the total number of fire counts as detected by the infrared channel of AVHRR on NOAA 11. The number of fire counts are separated by states: Pará, Tocantins, Mato Grosso, and Goiás, from June to November 1992. The sequence of fire events maximize by the end of August to the first 10 days of September, but in addition, the state of Pará shows a larger number of fires in October (for September 1992, see also the distribution by state shown by Kirchhoff and Alvalá

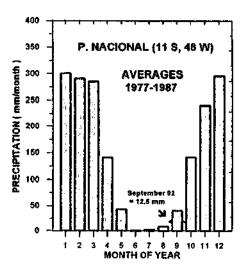


Figure 3. Precipitation at Porto Nacional, Tocantins, showing precipitation rates during September 1992 that are less than one half of the climatological average (1977–1987).

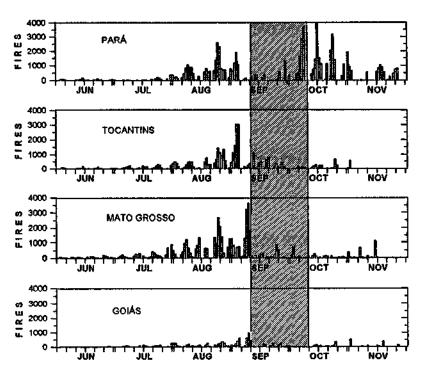


Figure 4. Total number of fire pixels from AVHRR for four Brazilian states in 1992. The hatched area represents the TRACE A period.

[this issue]. The remaining states, all in the cerrado area of central Brazil, show a much lower fire activity in the shaded area shown in Figure 4, which is the time period of the TRACE A mission in Brazil, from about September 10 to October 10, 1992. As mentioned above, the TRACE A field mission was actually planned to occur 15 days before it eventually did, but aircraft trouble caused a time delay of the mission, which otherwise would have occurred during the peak burning season.

## Results and Discussion Surface Ozone

ments that use the UV absorption technique are much simpler to make and have much better statistics since they are made almost continuously. Several interesting ozone features have been deduced from surface ozone observations [Kirchhoff, 1988; Kirchhoff and Rasmussen, 1990] and by making additional observations at a control station (outside of the burning area), it is possible to determine that part of the ozone concentration produced by biomass burning photochemistry, the excess ozone [Kirchhoff et al., 1992a]. Photochemical studies of interest have been described by Jacob and Wofsy [1988, 1990], Logan [1985], and Logan and Kirchhoff [1986] for ozone in situ measurements; by Thompson et al. [this issue], Pickering et al. [1992, this issue], and Scala et al. [1990] for the vertical redistribution of precursors; and by Watson et al. [1990], with satellite sensors. Surface ozone concentrations were measured

In comparison to ozone soundings, surface ozone measure-

#### Dry × Wet Season Results

Figure 5 shows ozone concentrations at Goiânia compared to the control station Natal, in September, part of the dry

routinely during TRACE A at the four sites of Figure 1:

Goiânia, Cuiabá, Porto Nacional, and Natal.

season period, and also during April, part of the wet season. Shown are the hourly averages for different months, with vertical bars which are typical standard deviations of the means and reflect the day-to-day variability of the observations. Average mixing ratios of 47 parts per billion by volume (pphv) are seen at 1600 LT at Goiánia, when the average mixing ratio at Natal is only 19 ppbv, an average excess ozone at Goiánia of 28 ppbv, in the dry season. These numbers are very similar also for October, not shown. In the wet season, however, Goiánia and Natal have very similar mixing ratios, as shown in Figure 5, right-hand panel. The difference of the two stations in the wet season period is 5 ppbv, at most.

The situation described above for Goiânia is very similar also at the other stations, Cuiabá and Porto Nacional. Figure 6 shows again two panels, one for the dry season period and one

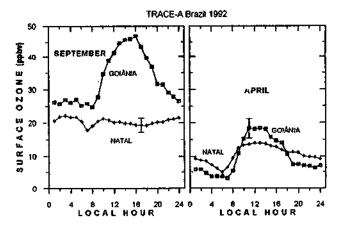


Figure 5. Surface ozone mixing ratios comparing results for the Goiânia and Natal sites during September and April 1992.

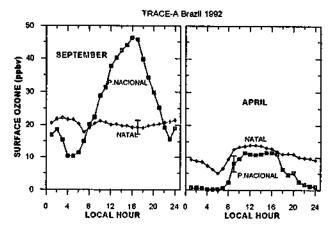


Figure 6. Surface ozone mixing ratios comparing results for the Porto Nacional and Natal sites, during September and April 1992.

for the wet season period, for Porto Nacional. Again, the daily maximum ozone mixing ratio at the surface occurs at 1600 LT, for the dry season period of September 1992, when an average of 47 ppbv ozone was observed in contrast to only 19 at Natal, an excess ozone of 28 ppbv (by coincidence, this number is equal to the one obtained for Goiânia). Again, the concentrations at Porto Nacional and Natal do not show much difference in the wet spason (April 1992), although the differences seem to be a little larger at Porto Nacional, compared to the Goiânia case. As in the previous figure, the vertical bars are the standard deviations of the means.

Average surface ozone mixing ratios at Cuiabá, the third observation site, during September, were a little smaller than those for the previous stations, 40 ppbv. This may be partly the result of the stronger precipitation activity near Cuiabá, during September 1992, which probably decreased the amount of nearby burning fuel. Evidently, the difference between Cuiabá and Porto Nacional at the time of maximum being only 7 ppbv, must also reflect the importance of air mass transport from neighbor source areas. Cuiabá often receives air masses from the north and west, where a larger percentage of deforestation takes place, with burning in the smoldering phase, which contributes larger amounts of CO (and therefore possibly also larger amounts of O<sub>3</sub>) in comparison to the flaming contributions of the Porto Nacional area.

#### Comparison With Previous Years

Cuiabá is one of the ground-based stations for which surface ozone measurements are available for previous years. Average surface ozone concentrations at Cuiabá (and Natal) for 1990, 1991, and 1992, shown in Figure 7, seem to follow the effect of the larger precipitation rates during 1992 at Cuiabá. The excess ozone exposure, defined by the shaded areas in Figure 7, is considerably smaller for the TRACE A 1992 period at Cuiabá. The case for Cuiabá illustrates the different behavior of observation sites in the same general ecosystem, the cerrado, but subject to different circulation systems.

## Ozone Soundings in the Troposphere Wet Season

From the previous surface ozone results at the Goiânia and Porto Nacional stations, shown in Figures 5 and 6, mixing ratios of less than 20 ppbv are expected near the surface, for the wet season. Figure 8 shows the ozone concentrations up to 100 hPa (16 km), the local tropopause, for April 13, 1992, at Goiânia. Three parameters are shown, the ozone mixing ratio in parts per billion by volume, the temperature T in Celsius, and the relative humidity in percent, RH. To illustrate the uncertainty of these measurements in the troposphere, for ozone, a "maximum uncertainty bar" is shown at 700 hPa, according to a comprehensive error analysis of Hilsenrath et al. [1986], who intercompared a number of different ozonesondes. Three different regions in the troposphere appear to be quite distinct. A surface layer, where the ozone mixing ratio increases with height, from about 10 ppbv near the surface to about 23 ppbv at 750 hPa. In this region the relative humidity (RH) is rather large, nearly 100%, and there is no temperature inversion visible in this case. Above 750 and below 300 hPa (where the relative humidity becomes negligible) the atmosphere seems to be well mixed in terms of ozone, the mixing ratio remaining almost constant at 23 ppbv over this height range. This seems to be the domain of most of the wet convection cells, but the humidity decreases sharply near 300 hPa, where apparently the third atmospheric layer starts. This is a very dry layer extending nearly up to 100 hPa, at which height level the ozone mixing ratio reaches out to the stratospheric values, above the tropopause, in this case in an abrupt manner. In this upper region there seems to be more vertical structure. The important new information that this case (and others, not discussed here) presents is that the ozone mixing ratio of this

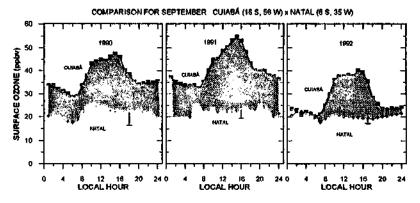


Figure 7. Excess ozone at the Cuiabá site for September 1990, 1991, and the TRACE A 1992 period.

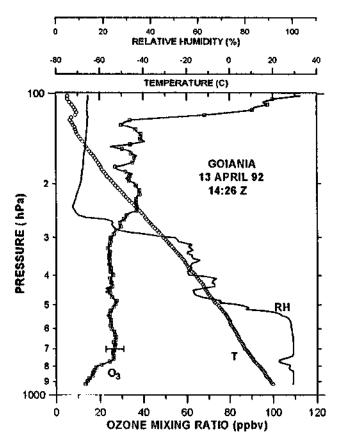


Figure 8. Ozone sounding for April 13, 1992, at the Goiânia site. Shown are the temperature (T), relative humidity (RH), and ozone  $(O_3)$ . The horizontal bar at 700 hPa for ozone indicates the maximum uncertainty of the measurement.

cerrado site remains low in the whole troposphere in the wet season, as was previously reported also for the Natal station [Kirchhoff et al., 1991]. The eight soundings made in April 1992, as indicated in Table 4, are shown as a mass plot in Figure 9. Mixing ratios for most of the soundings fall around 20-40 ppbv, with a small bulge near 300 hPa peaking at about 55 ppbv. All of these cases show the transition region troposphere-stratosphere near 150 hPa.

#### **Dry Season**

Despite the heavier rainfall at Cuiabá in 1992 compared to previous years, surface ozone in terms of monthly means was 40 ppby in September, which is an increase over the rainy season observed at Goiânia of about a factor of 2. This means that despite the unequal precipitation rates shown, the region is rather homogeneous in terms of surface concentrations of ozone. This is probably a consequence of the fact that rain is not the only factor that may affect surface ozone concentrations. Other important conditions may include wind directions and cloud cover.

For Cuiabá it can be seen that the ozone concentrations in the dry season are larger than the wet season values, in the whole troposphere. Figure 10 shows the results of the sounding of October 2 at Cuiabá. In comparison to the Goiánia case, the two lower atmospheric layers are much shallower. The initial region where the mixing ratio increases with height terminates near the 850-hPa level. The relative humidity falls to its lowest value just after the 600-hPa level, where a small temperature

inversion may be identified. Most evident in Figure 10 is the vertical structure, shown hatched, forming layers of higher ozone concentrations, similar to those reported by *Kirchhoff and Marinho* [1994]. In this figure, an arbitrary "background" ozone is set by connecting the lowest ozone mixing ratios from the ground to 200 hPa. The hatched ozone layers are then the ozone enhancements above this arbitrary background ozone.

The highest ozone concentrations, especially near the surface, were observed at Porto Nacional, probably because of the atmospheric conditions discussed earlier (drier than Cuiabá and Goiânia). Mixing ratios of more than 80 ppbv were seen on September 17, shown in Figure 11. The same parameters are shown, as in Figure 10 for Cuiabá. In this case, an enhanced ozone layer has been identified near 400 hPa. The difference between the driest site and the Goiânia site, where the wet season observations were made, is shown in Figure 12, where the ozone profiles for April 13 (Goiânia, one of the lower ozone concentrations) and September 19 (Porto Nacional, one of the larger ozone concentrations) are seen. It may be argued that the hatched area is an extension to the excess ozone, defined earlier for the surface, indicating that this parameter can remain the size of the surface value in the entire upper troposphere. Note also that near 800 hPa a narrow ozone layer is present at Porto Nacional, with peak concentrations of 100 ppby.

#### Variability in the Upper Troposphere

Figure 13 shows a mass plot at Porto Nacional for September 1992 (15 days; see Table 2) and Figure 14 shows the September results for Natal (11 days; see Table 3). There are two major differences, one near the surface and the other in the upper troposphere. For Porto Nacional near the surface there is a large scatter of points, as expected, reflecting the variability and intensity variation of nearby sources of ozone,

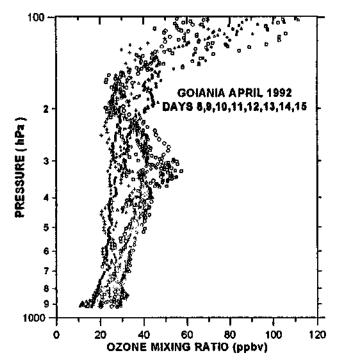


Figure 9. Mass plot of ozone mixing ratios for soundings at Goiânia, April 1992.

i.e., biomass burning events. For the 15 days of observation, ozone mixing ratios have varied between 20 and 90 ppbv near the surface and up to heights of 700-600 hPa, where the scatter in the data points starts to be smaller up to about 350 hPa. In contrast, for Natal, a station located quite far from sources, the variability near the surface is much smaller, between 20 and 40 ppby, and above the surface up to about 700 hPa clearly increasing the ozone mixing ratio with height, suggesting that the source of ozone is not near the surface, in contrast to the case for Porto Nacional. This suggests a source not on the ground but in the atmosphere itself. Trajectory analyses clearly indicate, at this low latitude, air mass transports from Africa [Pickering et al., this issue; Fuelberg et al., this issue; Thompson et al., this issue], thus suggesting that at Natal, ozone in the lower atmosphere is originated from African biomass burning products, transported over the Atlantic Ocean to Natal.

In the upper troposphere the scatter of data points is larger at Natal than at Porto Nacional. Considering that the number of data points is still limited, this result may be fortuitous, or it may be related to the fact that the ozone that is seen over Natal, in the upper troposphere, has its origin from distant (Brazilian) sources. Air mass transport over large distances and under very different circumstances brings these products to Natal, contrary to the case of Porto Nacional, which is directly in the source region. Photochemical calculations for the upper troposphere in the South Atlantic region, between about 8 and 100 m, show that this is an ozone source region in

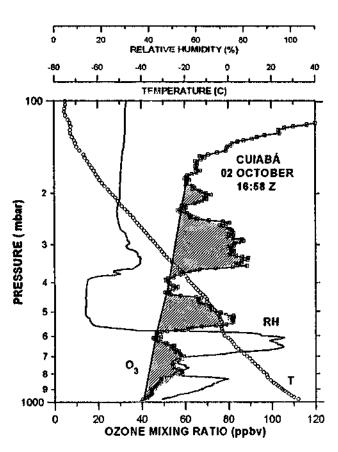


Figure 10. Ozone sounding for October 2, 1992, at Cuiabá, showing mixing ratios of ozone (and the indication of four major layers), temperature, and relative humidity.

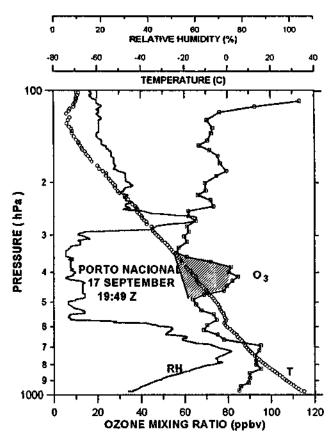


Figure 11. Ozone sounding for September 17, 1992, at Porto Nacional, showing again temperature, relative humidity, and ozone. The ozone mixing ratios are larger, and an ozone layer has been identified.

which photochemical production slightly exceeds loss [Jacob et al., this issue; D. J. Jacob, unpublished data, 1996]. Thus ozone is produced in this height region, and since ozone has a long lifetime here, it may be transported over large distances, from the source areas to Natal, and even farther, over the South Atlantic

A comparison of the results is shown in Figure 15, with averages of all measured profiles for the four sites. The horizontal bars are the standard deviations from the means, at the heights shown, and represent the variability of the ozone mixing ratios during TRACE A. The Goiânia (GO) profile has the smallest ozone mixing ratios, roughly around 30 ppbv, but with a clear indication of a vertical gradient which increases with height from the surface to 8 km. This characteristic is almost exactly identical for the other biomass burning sites, Cuiabá (CB) and Porto Nacional (PN). As mentioned before, the higher ozone mixing ratios expected for Porto Nacional, a site where the precipitation intensity for 1992 was lower than the climatological average, occurred not just near the surface, i.e., near the sources, but were spread out in the upper troposphere at concentrations near the 80 ppbv value. The ozone mixing ratio profile at Natal is different in this height region. There is also a region between the surface and an upper limit in which the ozone mixing ratio increases with height, but with two major differences: first, the vertical gradient is much stronger at Natal and, second, the upper height limit is much smaller than for the biomass burning sites, around 3.5 km. Above this limit, the Natal average mixing ratio during TRACE A is

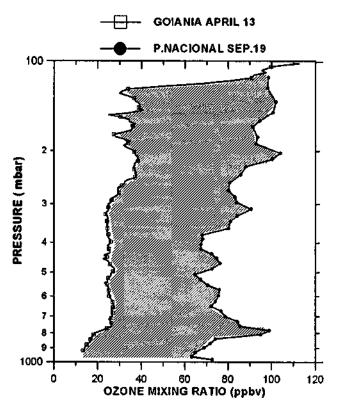


Figure 12. Variability of the ozone mixing ratio between a day of the wet season, with low ozone, and a day of the dry season, with relatively high ozone. The difference in the mixing ratio is roughly the same from the ground upward.

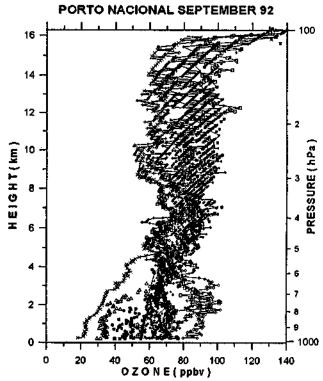


Figure 13. Mass plot of ozone mixing ratios for the Porto Nacional soundings.

slightly larger than that observed at Porto Nacional but is still within the standard deviations of the mean.

#### Comparison With Natal Ozone Climatology

It was shown before [Kirchhoff and Rasmussen, 1990] and again during TRACE A (Figures 5 and 6) that the surface observations of ozone are almost the same at Cuiabá and Natal, in the wet season. The new observations obtained during the wet season at Goiânia show that the ozone in the whole troposphere follows the behavior at the surface. It suggests that in the wet season the ozone concentration over the continent is not much different from the results near the coast, which in the lower troposphere at Natal represent air masses that come from the tropical South Atlantic. In Figure 16 the Goiânia result is compared to the climatological mean ozone in the wet season at Natal [Kirchhoff et al., 1991]. The Goiânia average (same as in Figure 15) is well within the standard deviations of the mean Natal (March, April, May, 1978-1988) values. It should be noted that if the Natal April average had been used for the comparison, the Natal values would be slightly larger than those shown in Figure 16, with about 12 ppbv more ozone near 200 hPa, 7 ppbv more near 500 hPa, and less than 2 ppbv more near the surface. This would not change the major conclusion derived from the result of Figure 16, namely, that a wet season data sample from the continent is comparable to the wet season climatology of the coastal site Natal, at the surface and in the whole troposphere. The April average for Natal, however, is much noisier than averages for either March or May, with standard deviations that are larger by factors of 2-4

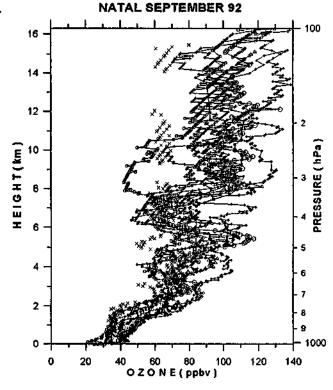


Figure 14. Mass plot of ozone mixing ratios for Natal soundings.

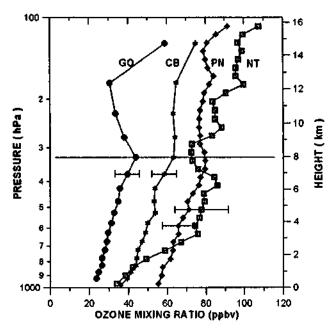


Figure 15. Average TRACE A profiles of ozone mixing ratios at the four sites of interest. The Goiânia (GO) average with eight profiles represents the wet season average. The other profiles were obtained in September-October, as described in the tables and text.

and therefore the average of Figure 16 seems to be more representative of the wet season characteristics.

The situation is somewhat different for the dry season, as shown in Figure 17, where the climatological Natal average is shown by the hatched area, which covers the length of the standard deviations of the Natal means. Also shown in Figure

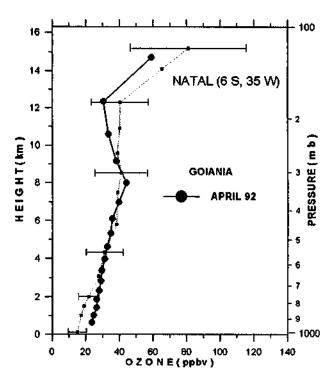


Figure 16. Comparison of the Natal climatological average for the local wet season with the April 1992 Goiânia average.

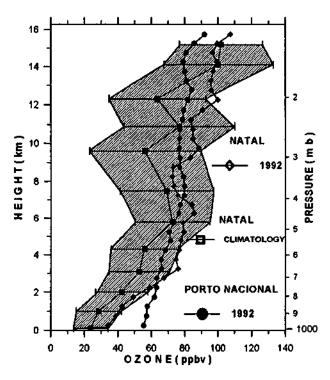


Figure 17. Comparison of the Natal September climatological average with the TRACE A site averages. See text.

17 is the Porto Nacional average 1992 profile (circles) and the Natal 1992 September-October profile (diamonds). Both 1992 profiles are within the standard deviations of the climatological Natal average but with slightly larger averages. The profiles are quite different in the lower 2 km, where Porto Nacional has clearly much larger ozone mixing ratios, with an average near surface value of 55 ppbv, whereas Natal has an average of about 31 ppbv, with a strong vertical gradient from the surface to about 3 km, above which height the ozone mixing ratios at the two stations are within the standard deviations of the Natal mean.

At Natal the vertical ozone concentration variability is dependent on the air mass origin from different sources, the most important of which are located thousands of kilometers to the west (the South American source) and tens of thousands of kilometers to the east (the African source). As shown by comprehensive trajectory analyses [Krishnamurti et al., this issue; Fuelberg et al., this issue], the sources to the west of Natal have important contributions to make at the higher levels of Natal, above about 500 hPa; they are also important beyond Natal, over Ascension Island, and the South Atlantic [Olson et al., this issue]. The data shown in this report suggest that the ozone concentrations at Natal, in the lower atmosphere, below about 500 hPa, have an African origin. This hypothesis was considered before [Kirchhoff and Nobre, 1986; Logan and Kirchhoff, 1986]. In the lower troposphere and at the surface the sources from Africa play a small but significant role in determining the ozone concentration at Natal, since the prevailing air mass trajectories in the lower troposphere travel in a "from east to west" direction [Shipham et al., 1993] according to different types of trajectory analyses [Harris and Kahl, 1990; Krishnamurti et al., 1993; Pickering et al., 1994]. On the contrary, in the upper troposphere, according to same references, the wind direction reverses and Natal receives air masses from the west, i.e., from the Brazilian tropical burning region.

#### Summary

#### Major Source Areas in Brazil

From close geographical analysis and year-by-year comparison of fire pixels from NOAA AVHRR, it is shown that the major source areas for biomass burning are presently the cerrado regions of central Brazil, plus the transition areas between the cerrado and the rain forest environment. In the first type of environment, flaming combustion of grass, shrubs, and small trees is the major biomass burning source. In the transition region between cerrado and forest, there is a larger contribution of smoldering combustion, which contributes larger quantities of reduced gases to the atmosphere. It has been shown on the basis of INPE deforestation studies that deforestations have decreased over recent years, and based on NOAA fire pixels, the major burning areas in Brazil presently maximize yearly in the north of the state of Tocantins, having two other large components, one near Marabá, in the state of Pará, and one in the north of the state of Mato Grosso, near the city of Alta Floresta.

## O<sub>3</sub> Concentrations in Source Regions Compared to Those Observed Outside

In the dry season the source regions represented by the three observation sites of this experiment have shown considerably more ozone than the control station, as expected. Considerable differences in precipitation rates at the three observation sites were observed, which complicates the analysis and conclusions of the comparisons. It appears that this difference in the precipitation regime in 1992 affected the ozone production in the lower atmosphere accordingly. It should be noted, however, that it is not just the precipitation itself that may be responsible for lower ozone concentrations, as a result of the decrease of the available fuel for burnings. Another important factor is the cloud cover, which sometimes exists over considerable areas without leading to rain but which may be responsible for less ozone formation, since cloud cover means absence or lower intensities of UV radiation, needed in the ozone-producing process. During this experiment, the surface ozone observations have shown hourly concentration averages of 47 ppbv at Porto Nacional and Goiânia and 40 ppbv at Cuiabá for the September averages observed at 1600 LT. From these average values and taking also into account the ozone values observed at Natal, the excess ozone of the source region, i.e., the ozone amount that one might associate directly to biomass burning, was of the order of 30 poby. This is a large amount of ozone if one considers that natural processes in the rainy season produce only about 15 ppby. Much larger values have also been observed occasionally. For example, at Porto Nacional, more than 80 ppbv have been observed near the surface and in the lower troposphere. The vertical ozone concentration profiles at Porto Nacional were also larger in comparison to those at Cuiabá. No significant ozone concentration differences have been observed in the wet season (April) between the Goiânia data sample and the Natal observations.

## Concentration Differences Among Biomass Burning Sites in the Same Seasonal Period

In the wet season, observed differences are negligible. The observations in the dry season were not totally adequate to

answer this question, since the precipitation rates at the sites were quite different during 1992. The results of this experiment indicate, however, a remarkable homogeneity in terms of ozone concentrations near the surface, with only small differences; drier sites (or sunnier sites) had slightly larger ozone concentrations in the lower atmosphere.

#### Upper Tropospheric Ozone and Surface Ozone

There is a tendency for tropospheric ozone (lower and upper troposphere) to follow the surface ozone variations, at time-scales of days. In the seasonal periods when surface ozone concentrations are small (large), the upper tropospheric ozone concentrations are also small (large).

#### Comparison of 1992 Observations With Previous Years

From surface ozone measurements of previous years at Cuiabá in September it is shown that ozone concentrations for the years 1991 and 1990 were larger by about 15 and 8 ppbv, respectively. Results for more recent years (1993, 1994, not shown) are also larger.

## Comparison of the Source Region O<sub>3</sub> Profiles With Natal Climatology in Wet and Dry Seasons

Our results confirm that in the wet season there are no significant differences between the profiles taken over the continent and at the coast. In the dry season the Porto Nacional average profile is within the standard deviations of the Natal climatological mean in the upper troposphere. This is an interesting new result, since one might have expected larger ozone concentrations in the source region profile compared to the coastal profile (which is true, however, in the lower atmosphere). This appears to be the result of larger production rates (production larger than loss rates) in the higher layers and a time constant of ozone in the upper troposphere of up to 4 weeks [D. J. Jacob et al., unpublished, 1996; Thompson et al., this issue]. This makes the ozone concentrations at these levels rather uniform over large distances, along the prevailing wind streamlines which have their general origin over the source area of central Brazil. The burning products in the upper layers are transported to and beyond Natal, over Ascension Island and into the South Atlantic [Pickering et al., this issue; Fuelberg et al., this issue]. The situation in the lower troposphere is different: it reflects a slight excess loss rate [Jacob et al., this issue; D. J. Jacob et al., unpublished, 1996] burning products of different origins and the existence of nearby sources for Porto Nacional, for example, with large values and concentration variations in the lower troposphere and at the surface. At Natal, trajectory analyses below 600 hPa show that the burning products originate from the Atlantic (thus ultimately from Africa) causing increasing concentrations from the surface up, with relatively small variability.

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