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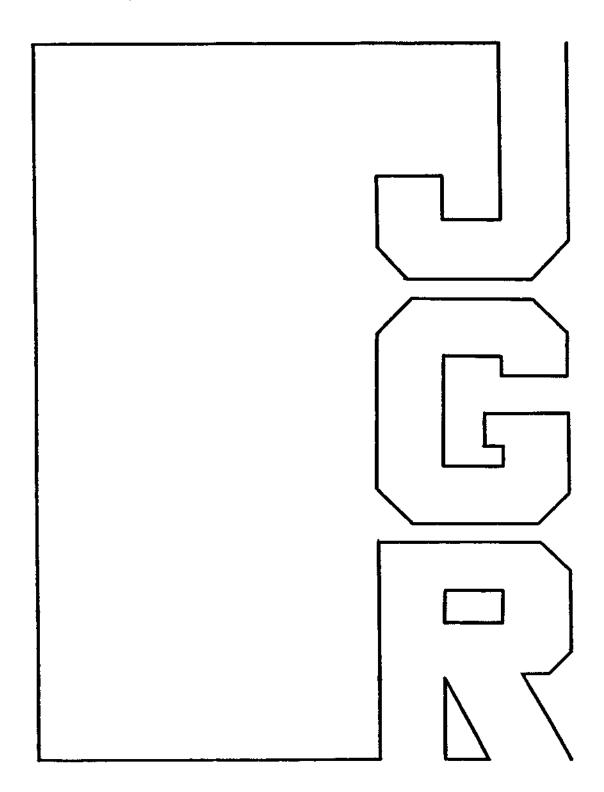
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Ozone and Aerosol Distributions Over the Amazon Basin During the Wet Season

E. V. Browell, G. L. Gregory, R. C. Harriss, and V. W. J. H. Kirchhoff



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Ozone and Aerosol Distributions Over the Amazon Basin During the Wet Season

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Measurements of ozone (O₃) and aerosols were made over the tropical rain forest of Brazil during the wet season in April-May 1987 as part of the NASA Global Tropospheric Experiment to study the Amazon boundary layer. Remote and in situ measurements of O₃ and aerosols were made from aircraft on flights over Brazil in the vicinity of Manaus and between Manaus and Belem. Ozonesonde data were also obtained near Manaus. Ozone mixing ratios of <12 ppbv were found in the mixed layer during the wet season with no significant evidence of O₃ produced from biomass burning or photochemistry. These values are lower than those found during the 1985 dry season by 6-8 ppbv. These low O₃ mixing ratios indicate a strong removal process near the surface during the wet season. The region from the mixed layer top to 3 km in altitude had a slowly increasing O₃ profile from 12 to 20 ppbv. On long-range flights between Manaus and Belem, no significant difference was found in the distribution of O₃ above the mixed layer between the inland tropical rain forest and the marine conditions near the coast. Within the mixed layer, there was a definite trend to lower O₃ levels above the forest compared to over the ocean. This reflects the marked difference in the sinks for O₃ over these two regions. The rate of growth of the mixed layer over the rain forest in the wet season was found to be ~ 9 cm s⁻¹, which is within the 7–10 cm s⁻¹ range found for the dry season. There was no evidence of the trade wind inversion that was seen during the dry season, and due to frequent precipitation, the background aerosol loading was lower in the wet season than in the dry season.

Introduction

Tropical rain forests represent a significant percentage of the land area of the world (6.6%) [Ajtay et al., 1979], and their interaction with the atmosphere permits the exchange of large quantities of gases and aerosols that can alter the composition and chemistry of the troposphere [e.g., Crutzen and Gidel, 1983; Crutzen, 1986]. In particular, the budget of O₃ in the troposphere over the rain forest can be directly affected by the vegetation acting as a surface for heterogeneous reactions that destroy O3 and by the emission of carbon monoxide (CO) and hydrocarbons that can react photochemically with other species to produce or destroy O₃, depending on the ambient concentrations of key gases such as nitric oxide (NO) [Crutzen et al., 1979, 1985; Jacob and Wofsy, 1988; Fan et al., this issue; Jacob and Wofsy, this issue]. We also know that the products of biomass burning can provide a large source of aerosols and the necessary components for the photochemical production of O₃ [Crutzen et al., 1985; Delany et al., 1985; Fishman et al., 1986; Fishman and Larsen, 1987]. These plumes can then be transported over long distances in the middle troposphere [Delany et al., 1985; Andreae et al., 1988] and eventually impact the global troposphere [Fishman, 1988].

To examine the various sources and sinks for gases and

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aerosols across the Amazon Basin, combined aircraft and ground-based field experiments were conducted in July-August 1985 to investigate the atmosphere over the rain forest during the "dry" season [Harriss et al., 1988] and in April-May 1987 to investigate the "wet" season [Harriss et al., this issue (a)]. Investigations of the variability of O₃ and aerosols over the tropical rain forest during the dry season were conducted as part of the 1985 NASA Global Tropospheric Experiment (GTE)/Amazon Boundary Layer Experiment (ABLE 2A), and the results from these investigations have been reported by Browell et al. [1988], Gregory et al. [1988], and Kirchhoff et al. [1988]. The knowledge gained during the ABLE 2A dry season field experiment provides an important reference for the results discussed in this paper for the wet season.

The wet season field experiment (GTE/ABLE 2B) consisted of an extensive set of surface (land and water), tower, balloon, and aircraft investigations [Harriss et al., this issue (a); Garstang et al., 1990]. This paper presents the results of O₃ and aerosol measurements made in the lower troposphere from April 13 to May 10, 1987, as part of this experiment. Aircraft flights on the NASA Electra were conducted in the vicinity of Manaus, Brazil, (3°S, 60°W), and between Manaus and the mouth of the Amazon River (0°S, 48°W). Airborne O₃ measurements were made in situ and with a lidar system pointed below the aircraft in a nadir mode of operation. Ozonesondes were launched in the vicinity of Manaus to investigate O₃ distributions across the tropo-

sphere and into the lower stratosphere. Aerosol distributions were measured in situ on the aircraft and above and below the aircraft with the lidar system. This paper discusses the distributions of O₃ and aerosols observed during the wet season from within the mixed layer to the middle troposphere over the central Amazon Basin and between Manaus and Belem, and these observations are compared to the measurements obtained in the 1985 dry season. Companion papers in this issue discuss the temporal and spatial variability of O₃ in the mixed layer across the Amazon Basin [Gregory et al., this issue], from the surface to the tropopause near Manaus [Kirchhoff et al., this issue], in and immediately above the rain forest [Fan et al., this issue], and in cloud transport studies [Scala et al., this issue].

AIRBORNE LIDAR AND IN SITU INSTRUMENTATION

The remote measurements of O₃ and aerosol distributions were obtained by an airborne differential absorption lidar (DIAL) system [Browell et al., 1983; Browell, 1989]. This system was configured to measure O₃ profiles along its vertical line of sight beneath the Electra aircraft. When in this nadir mode of operation during the daytime, O_3 mixing ratios can be obtained with the DIAL system from a range of 750 m below the aircraft to about 300 m above the forest canopy with an accuracy of <10% and a vertical and horizontal resolution of 210 m and 6 km (300 laser shots), respectively [Browell, 1983; Browell et al., 1983, 1985a, b]. Prior to this field experiment, data from this system have been used in numerous studies of tropospheric O₃ and aerosols [Browell et al., 1983, 1985b, 1987, 1988; Shipley et al., 1984; Fishman et al., 1985; Vukovich et al., 1985; Gregory et al., 1986, 1988; Andreae et al., 1988; Ching et al., 1988; Fishman and Browell, 1988; Garstang et al., 1988; Kirchhoff et al., 1988; Martin et al., 1988]. The two laser pulses used in the DIAL measurement of O₃ in this study were independently tuned to a wavelength that is strongly absorbed by O₃ (286 nm) and to a wavelength that is only slightly absorbed by O₃ (300 nm). The output pulses were transmitted nearly simultaneously ($<300 \mu s$ separation) in a traditional lidar mode to obtain range-dependent atmospheric scattering and absorption information at the two ultraviolet (UV) wavelengths. The relative change in the two lidar signals between any two ranges is predominantly due to the absorption of O₃. This relative change can be related to the average O₃ concentration between the two ranges. An O₃ profile can then be derived by calculating the absorption over 210-m range intervals along the UV lidar returns. Additional details on the airborne DIAL system and the technique used in the DIAL O₃ data reduction are discussed by Browell et al. [1983, 1985a] and Browell [1989]. The raw lidar returns were digitized with a vertical resolution of 15 m, and the laser pulses were transmitted at 5 Hz, which represents a horizontal spacing between the lidar returns of about 20 m. These data were averaged vertically and horizontally to reduce the DIAL O3 measurement statistics.

Laser pulses at 600 and 1064 nm were transmitted above and below the aircraft at the same time that the UV laser pulses were being transmitted below the aircraft. These measurements were also made at a 5-Hz rate; however, because of the better statistics associated with the measurement of aerosol distributions at these wavelengths, the

vertical and horizontal resolution of the measurement could be as small as that defined by the digitization rate of the lidar return (15 m) and the laser pulse rate (\sim 20 m), respectively. The use of lidar returns at 1064 nm provides good contrast between molecular and aerosol backscattering and less atmospheric attenuation than at shorter wavelengths. This results in more sensitive detection of atmospheric aerosol distributions along the lidar line of sight. Continuous color plots of O_3 mixing ratios and aerosol backscattering as a function of altitude and distance along the aircraft ground track were obtained in real time on the aircraft for selection of in situ sampling altitudes or spiral locations.

Two chemiluminescence O₃ systems were used for continuous in situ measurements on the aircraft [Gregory et al., 1983, 1988, this issue]. The in situ O₃ data used in this study were from an ethylene (C2H4) chemiluminescence instrument which has an O3 mixing ratio measurement accuracy of 5 ppbv (parts per billion by volume) or 5% (whichever is larger) with a 2-s response time. Aerosol number densities in the 0.12- to 3- μ m diameter range were measured by an active scattering spectrometer probe (ASASP). The ASASP counts and sizes aerosols in 15 bins with bin widths progressively increasing from 0.025 to 0.375 μ m for aerosol diameters from 0.12 to 3 μ m, respectively. Aerosol counts are typically averaged for 10 s for reporting purposes. Total air temperature was measured on the aircraft with a platinum resistance probe. The probe has a measurement accuracy of ±0.5°C and a response time of 1 s. Dew point temperature was determined with a cooled-mirror hygrometer, which has a measurement accuracy of ±0.2°C and a response time of 2°C s⁻¹. Temperature and dew point measurements were used to provide information on the characteristics of layers observed in the atmosphere. Ozone profiles in the upper troposphere and lower stratosphere (≥10 mbar) were obtained from 20 electrochemical concentration cell (ECC) sondes [Komhyr, 1969; Barnes et al., 1985; Kirchhoff et al., 1988] launched near Manaus over the period from April 16 to May 10, 1987 [Kirchhoff et al., this issue].

The NASA Wallops Flight Center Electra aircraft was used as the platform for all flights in this field experiment. It has a nominal air speed of 120 m s^{-1} and a maximum flight endurance of 6.5 hours (depending on instrument load). The ceiling for this aircraft is about 9 km; however, these experiments did not require flight altitudes above 5 km. In addition to the above measurements, the Electra had instrumentation for in situ measurements of CO, NO, carbon dioxide, methane, nonmethane hydrocarbons, sulfur, radon, peroxyacetyl nitrate, organic acids, aerosol composition, and fluxes of O_3 , CO, and water vapor. A general description of these systems and their measurements is given by *Harriss et al.* [this issue (a)], and more details are provided in the other papers in this issue.

DATA RESULTS AND DISCUSSION

Ozone and aerosol data were obtained on 19 aircraft flights over the Amazon Basin between April 13 and May 8, 1987. The locations of these flights are shown in Figure 1, and a list of the missions is given in Table 1. The "source," "transport," and "flux" flights were made within about 400 km of Manaus, and the "survey" flights covered about 1500 km between Manaus and the coastal Atlantic near the mouth of the Amazon River. The source flights investigated the natu-

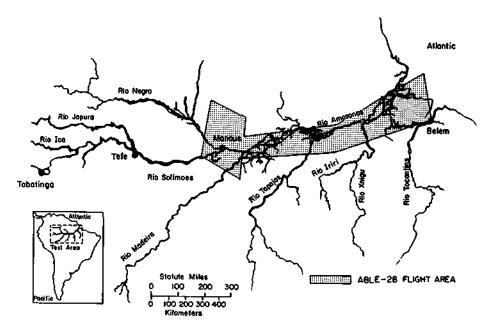


Fig. 1. Map of regions investigated for O₃ and aerosol distributions during the GTE/ABLE 2B field experiment.

ral source (or sink) of gases and aerosols over the rain forest, and they involved repeated flights at different altitudes (0.2-5 km) between fixed locations starting soon after sunrise and continuing into the early afternoon. This provided an opportunity to study the temporal development of the conditions over the rain forest. One only nighttime flight was made to examine the shallow nocturnal mixed layer and stable free troposphere over the same area flown during the daytime missions. Transport studies associated with convective storms were conducted in two modes: (1) a series of measurements were made along an atmospheric cross section (or "wall") ahead of and, when possible, behind a line of storms, and (2) flights were made around a well-instrumented mesoscale triangle at different altitudes to study the

divergence/convergence of the air associated with the storm. The results from these convective transport studies are presented in the work by Garstang et al. [1990] and Scala et al. [this issue]. The survey missions were composed of a series of high-altitude (~3 km) and low-altitude (150 m) legs that were 75-200 km in length. Details of the flights conducted during the ABLE 2B field experiments are given by Harriss et al. [this issue (a)].

Ozone and Aerosol Observations Over the Central Amazon Basin

Undisturbed daytime conditions. Atmospheric cross sections of aerosol and O₃ distributions obtained with the airborne DIAL system during ABLE 2B under typical day-

TABLE 1. Summary of ABLE 2B Missions

Date in 1987	Mission Type	Flight Times, UT		Flight
		Takeoff	Landing	Altitude Range, km
April 13	source	1743	2226	0.2-3.0
April 14	source	1656	2231	0.2-4.3
April 15	transport-double wall	1604	2132	0.2-4.6
April 17	transport-volume	1331	1920	0.2-4.6
April 19	source	1352	1918	0.2-4.6
April 20	source	1605	2143	0.2-4.6
April 21	transport-volume	1355	1945	0.2-4.6
April 23	Manaus to Belem survey	1054	1622	0.2-3.0
April 24	Belem to Santarem survey	1107	1547	0.2-3.0
April 24	Santarem to Manaus survey	1716	2129	0.2-3.0
April 26	source	1057	1505	0.2-3.0
April 28	source	1121	1650	0.2-3.0
April 29	source-forest	1159	1737	0.5-4.3
April 30*	source-night	2202	0312	0.2-3.0
May 2	source-wetlands	1524	2049	0.2-4.3
May 4	flux	1502	2026	0.8-3.0
May 6	transport-volume	1254	1826	0.2-3.7
May 7	source	1313	1453	0.2-5.5
May 8	transport-single wall	1537	2104	0.2-4.6

Universal time, or GMT, is equal to Manaus time plus 4 hours.

*Takeoff date.

time conditions are given in Plate 1. These data were obtained on May 2, 1987, during the afternoon and in the absence of any major storm activity in the immediate vicinity of the aircraft. It should be noted that localized, widely scattered showers were observed on most afternoons during our flights. The false color displays in Plate 1 of the relative aerosol backscattering and O3 mixing ratio distributions were obtained with the airborne DIAL system operating in a nadir mode from the Electra aircraft at an altitude of 4.2 km (unless otherwise noted, all altitudes in this paper are geometric altitudes above ground level). The top display represents the aerosol distribution derived from the 1064-nm lidar data. Each vertical line represents an average of 15 lidar returns over about 300 m horizontally with a vertical resolution of 15 m. The solar background and range-squared dependence of the lidar signal were removed prior to producing this display, and because of the high-visibility conditions that existed throughout the experiment (outside of clouds), no correction for atmospheric attenuation was needed in the analysis. The top of the forest canopy can be seen as a black line at the bottom of the display. More aerosol scattering is indicated by the yellow, orange, and red colors, and black represents very highly scattering aerosols and clouds. Magenta and blue colors represent low amounts of aerosol scattering in the absence of strong aerosol attenuation (e.g., near 1930 UT), and white represents a region of no data, such as under an optically thick cloud or during a time gap in the data due to an aircraft turn. Since the relative humidity in the lower troposphere (≤3 km) was found to generally exceed 70% during ABLE 2B, aerosol scattering was greatly increased because of the hygroscopic growth of the aerosols at these high humidities. It is difficult to quantitatively interpret the lidar aerosol data in terms of aerosol loading, since the amount of scattering is dependent upon a combination of aerosol abundance, size distribution, and relative humidity effects. Much of the horizontal inhomogeneity that is present in the aerosol data can be attributed to inhomogeneity in relative humidity and not specifically to aerosol number density changes. Since the lidar backscattering at 1064 nm is most sensitive to aerosols with diameters of $>0.5 \mu m$, the growth of aerosols due to hygroscopic effects makes a significant impact on the aerosol backscattering data. The primary source of the aerosols and water vapor is from the rain forest, and thus the regions that are observed with the lidar to have enhanced aerosol scattering are associated with air in the mixed layer or air that has been transported out of the mixed layer by convective mixing processes.

The top of the mixed layer is identified at the first abrupt decrease in aerosol scattering that occurs at the top of the layer that is in contact with the ground. This estimate is made away from clouds, since clouds will often penetrate the weak temperature-stable region at the top of the mixed layer to rise to a higher stable region. This technique for estimating the mixed layer top can be seen to be relatively unambiguous at the two ends of this cross section; however, this technique becomes difficult to apply when there are strong aerosol scattering layers directly above the mixed layer, as can be seen in the center of this plate. The average depth of the mixed layer across this atmospheric cross section is estimated to be about 800 m above the forest canopy. Convective plumes from surface heating effects can be seen at the top of the mixed layer on the right side of the figure.

Clouds at many different levels can also be seen in this cross section. The atmosphere is so conditionally unstable during the wet season that the release of latent heat in clouds provides enough buoyancy to allow the clouds to rise into the middle or upper troposphere and possibly initiate a local storm event. The region above about 2.5 km has less aerosol backscatter than at lower altitudes, and we know from the in situ measurements that at this altitude the relatively humidity was less than 60% and that the aerosol loading was very low.

The O₃ mixing ratio distribution shown at the bottom of Plate I was obtained with the airborne DIAL system over the same region represented by the aerosol scattering cross section. Each vertical line represents the average of 300 lidar returns (1-min average or about 7-km horizontal distance) with a 210-m vertical resolution. A horizontal running average was used to present a continuous O3 plot that was in phase with the aerosol data. The lidar data start about 750 m below the aircraft, and due to the 210-m smoothing interval, O₃ DIAL calculations are not made within about 300 m of the surface (canopy top). White areas represent data gaps due to clouds, aircraft turns, or instrument reasons. The color scale for O₃ at the top of the O₃ color plot is the same as that discussed previously for the relative aerosol scattering cross section. The color scale covers the range of O₃ mixing ratios from zero to 50 ppbv. Two trends are readily identified in the O3 distribution. First, in the altitude range from 2 to 3.2 km, O₃ has an increasing trend from about 16 to 20 ppbv along this flight track; and second, O3 decreases below 2 km to an average of about 10 ppbv at 500 m. There are clearly lower O3 mixing ratios within the mixed layer than above it. The region from the top of the mixed layer to 2 km is a transition region for O₃ from the low levels in the mixed layer to the higher, more uniform levels above 2 km. The transition region can also be seen in the aerosol scattering distribution at the top of Plate 1. Since aerosols and moisture are derived from the surface and are thus enhanced in the mixed layer, the high degree of inhomogeneity in aerosols and moisture in the transition region reflects the high spatial variability in mixing of mixed layer air and the drier, cleaner air aloft.

There was typically less than 15% variation in the average O₃ profiles above the mixed layer during the wet season in the absence of a major storm; however, Plate 2 presents the DIAL data for the case of the largest change in O₃ and aerosol distributions observed during this experiment. On this day, the flight area was under the influence of strong atmospheric subsidence, which prevent cloud transport above 2.5-3 km. The convective clouds were formed near the top of the mixed layer, and some of them ascended into the relatively stable region above the mixed layer. The O₃ distribution shows unusually elevated O3 mixing ratios (20-30 ppbv) above 1.5 km. The cloud density prevented DIAL O_3 measurements within the mixed layer (<800 m), but the O_3 in the transition region from ~ 0.8 to 1.5 km was also found to be slightly higher than normal. This example has the highest O₃ mixing ratios observed with the airborne DIAL system during the wet season.

The average O_3 profiles for the atmospheric cross sections shown in Plates 1 and 2 are plotted in Figure 2 along with the average in situ O_3 measurements made at 0.15 and 4.2 km. The O_3 profile obtained on April 29, 1987, was found to be 7-10 ppbv higher than the O_3 profile measured on May 2,

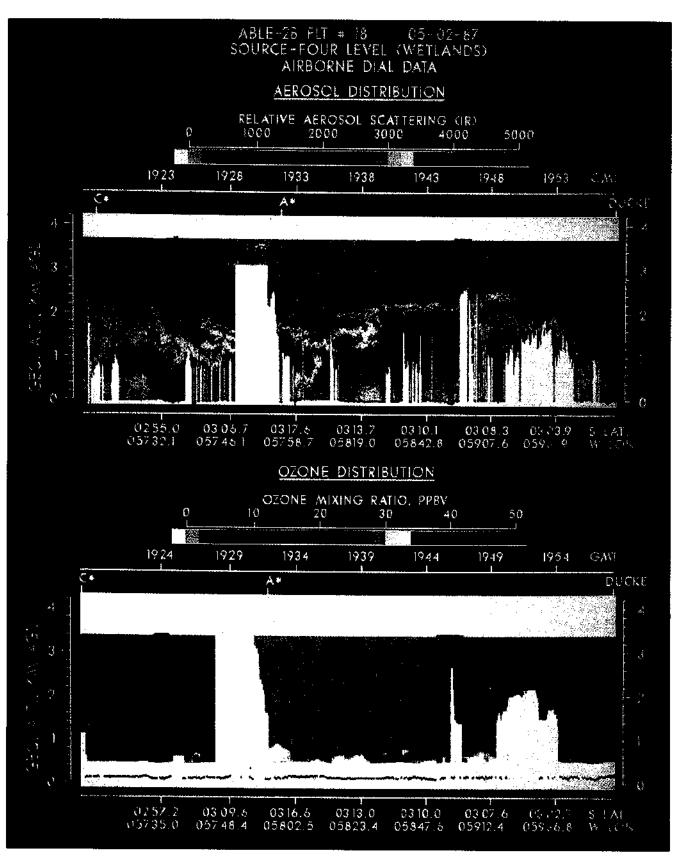


Plate 1. Aerosol and O_3 distributions obtained during the afternoon of May 2, 1987, in the absence of any major storm activity. (Top) Airborne DIAL aerosol data are shown in a false color display with the relative amount of atmospheric backscattering defined in the color scale given at the top of the display. (Bottom) The airborne DIAL O_3 data are also shown in a false color display with the O_3 mixing ratios in parts per billion by volume (ppbv) defined in the color scale at the top of the O_3 display. In both cases, black represents values greater than the maximum level given on the color scale. Geometric altitudes are given in kilometers above ground level (AGL) or above the top of the forest canopy; Greenwich mean time (GMT) or universal time (UT) is shown at the top of each display (Manaus time = GMT – 4 hours); and aircraft latitude and longitude information at each reference time is given at the bottom of each display (latitude and longitude are given in degrees and minutes, e.g., 0250.3 is 2°50.3'). The aircraft altitude for these measurements was 4.2 km.

overpasses of the same general area during the morning. The observed growth rate of $9.4 \,\mathrm{cm\ s^{-1}}$ is comparable to the 7-10 cm s⁻¹ rate found during the dry season [Browell et al., 1988]. The development of clouds and the effects of precipitation and the vertical transport of air in the vicinity of deep convective storms were clearly evident in the lidar aerosol data. In a few cases, air from above the mixed layer with higher O_3 and lower aerosol loading compared to air in the mixed layer was observed to be transported down to near the surface in the vicinity of a convective storm. Even with the relatively small vertical gradient in the background O_3 profile, airborne DIAL O_3 profiles have been used by Garstang et al. [1990] and Scala et al. [this issue] to study vertical convective transport during the wet season.

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