# Transport of ozone to the surface by convective downdrafts at night

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[1] During the Large-Scale Biosphere-Atmosphere Experiment in Amazonia wet season experiment, the near-surface measurements of equivalent potential temperature and ozone at night, when background levels of ozone are low, clearly show that convective downdrafts rapidly transport air with higher ozone and lower equivalent potential temperature down to the surface from around 800 hPa. This largely unreported downward transport of ozone may play a significant role in the photochemistry of the atmosphere boundary layer and increase the surface deposition of ozone. *Index Terms:* 0368 Atmospheric Composition and Structure: Troposhere-constituent transport and chemistry; 3314 Meteorology and Atmospheric Dynamics: Convective processes; 3307 Meteorology and Atmospheric Dynamics: Boundary layer processes; 3374 Meteorology and Atmospheric Dynamics: Tropical meteorology
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## 1. Introduction

[2] The Wet Season Atmospheric Mesoscale Campaign (WETAMC) of the Large-Scale Biosphere-Atmosphere (LBA) Experiment in Amazonia in January and February 1999 afforded an excellent opportunity to study the interaction of the boundary layer (BL) with convection over land in the deep tropics [*Silva Dias et al.*, 2000]. At one pasture site at Ouro Preto d'Oeste in Rondônia (10°46.42′S, 62°20.22′W) a surface mesonet site and flux tower measured the diurnal thermodynamic cycle and the surface energy balance, while tethered balloon and rawinsonde ascents probed the atmospheric layer above. The site is part of a large deforested area (>250 000 km$^2$) dominated by a short grass (*Brachiaria brizantha*) with isolated palm trees scattered throughout the landscape. The pasture is a rural, nonpristine site, with a well-traveled highway within 10 km to the northeast. The landscape surrounding the measurement site is reasonably flat and located south of the forested regions of Amazonia. At the same time as the meteorological measurements, the concentration of trace gases (ozone ($O_3$), carbon monoxide (CO), $NO_x$ (= nitric oxide (NO) + nitrogen dioxide ($NO_2$)), and volatile organic compounds (VOCs)) and aerosol mass were measured [*Gatti et al.*, 2000]. In this paper we use nighttime data to show that convective downdrafts, which bring down air of lower equivalent potential temperature, $\theta_E$, to the surface from the lower-middle troposphere, also bring down air of higher ozone concentration. We shall first discuss the $\theta_E$ structure of the tropical atmosphere and then review briefly the processes that affect ozone concentrations in the tropical boundary layer.

[3] The study of moist convective processes and transports have long used $\theta_E$ as a tracer, because it is conserved in the condensation and evaporation of water [e.g., *Emanuel*, 1994]. The primary source of $\theta_E$ is at the surface, where the surface sensible and latent heat fluxes, driven by solar heating, can be considered a $\theta_E$ source to the atmosphere [e.g., *Betts*, 1992], and the primary sink is radiative cooling of the troposphere. Consequently, the tropical atmosphere is characterized by a decrease of $\theta_E$ with height. The tropical atmosphere is always close thermally to moist neutrality [e.g., *Riehl*, 1979; *Betts*, 1998], so the vertical transports by moist convection are the primary mechanism for the vertical mixing of the atmosphere. The ascent path of air parcels ascending in clouds follows a moist adiabat, a path of constant $\theta_E$ (and so transport high $\theta_E$ air upward from the near-surface mixed layer), while the evaporation of falling rain into unsaturated air also conserves $\theta_E$ to good approximation, and the cooled air sinks in downdrafts, which bring low $\theta_E$ air down from the lower-middle troposphere into the boundary layer (BL), and to the surface. Many papers have used $\theta_E$ as a tracer to track downdraft air [e.g., *Zipser*, 1969; *Betts*, 1973, 1976; *Betts and Silva Dias*, 1979]. For this analysis we computed $\theta_E$ from 1-min average micrometeorological measurements of pressure, temperature, and humidity on a tower at a height of 1.5 m [*Betts et al.*, 2002] at this pasture site and from radiosondes launched from the site.

[4] Although ozone in the atmospheric boundary layer exists only in the parts per billion (ppb) range, it is an important gas due to its key role in influencing the oxidation capacity of the lower atmosphere. Without ozone, chemical species such as CO and $NO_x$, which are introduced to the atmosphere due to anthropogenic and biogenic activity, could accumulate in the lower atmosphere to harmful levels. Conversely, tropospheric elevated ozone levels can contribute to altering Earth's surface energy balance, as ozone strongly absorbs thermal en-

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ergy in the so-called atmospheric window (in the wavelength of ~9.6 $\mu$m). In the unpolluted free troposphere, ozone levels are relatively high compared with those at the surface, as stratospheric ozone can be transported downward into the upper troposphere [*Kirchhoff et al.*, 1990]. Near the surface in the atmospheric boundary layer, it can be both generated and removed by catalytic and photochemical reactions involving CO, VOCs, and $NO_x$. Despite the extensive research carried out during the last decade [*Brasseur et al.*, 1999; *Sachse et al.*, 1999], we still do not know the exact contribution of the stratosphere to the ozone levels recorded close to the ground. Since the tropopause is much higher in the tropics than in midlatitudes, it has been suggested that much of the ozone observed in the rural lower atmosphere is photochemically produced [*Crutzen*, 1985], because processes such as natural fires and biomass burning, in general, contribute to the production of precursors which form ozone [*Pickering et al.*, 1992; *Thompson et al.*, 1997]. Studies have shown that near-surface photochemical generation is large during the dry season, in part because of widespread biomass burning [*Delany et al.*, 1985; *Kirchhoff et al.*, 1990]. In the rainy season, ozone production in Amazonia is reduced, because of the suppressed photochemical activity resulting from somewhat cloudier conditions (and hence less actinic irradiance to drive ozone photochemistry) and from little or no biomass burning. Additionally, the dry deposition of surface ozone to vegetation is enhanced during the rainy season [*Gregory et al.*, 1988; *Fan et al.*, 1990; *Sigler et al.*, 2001]. Consequently, in the rainy season the Amazonian region is predominantly a sink of ozone, and ozone concentrations increase with height [*Gregory et al.*, 1988]. During the daytime, when vertical mixing is strong in the BL, surface ozone values are high in response to local photochemistry and vertical transport from aloft to the surface. At night, as the surface cools and the stable BL largely uncouples the surface from the troposphere above, near-surface values become low because of dry deposition of ozone to the underlying surface [*Kirchhoff*, 1988; *Fan et al.*, 1990]. As a result, ozone levels progressively decline after sunset, and within a few hours the nocturnal BL generally becomes low in ozone due to surface deposition. In earlier field and modeling investigations in the tropics [*Garstang et al.*, 1988; *Scala et al.*, 1990] the emphasis was to examine the upward transport of trace gases (including ozone) from the surface into the free troposphere by convective systems. In contrast, in this study we examine the largely unreported vertical transport of ozone from the middle troposphere to the ground surface during highly convective storm systems. In the presence of convective downdrafts, driven by the evaporation of falling precipitation, ozone levels can rapidly increase at night due to the strong coupling between the surface and the air aloft. We shall show examples where these downdrafts penetrate the stable BL at night and inject air from above, that is both low in equivalent potential temperature and high in ozone.

## 2. Ozone and Equivalent Potential Temperature Variations at the Surface

[5] The coupling of ozone ($O_3$) and equivalent potential temperature ($\theta_E$) is different during the daytime period, while the convective boundary layer (BL) is deepening under the influence of surface heating, and at night, when mean ozone is low unless convective downdrafts bring ozone down to the surface. We shall first show the mean diurnal cycle.

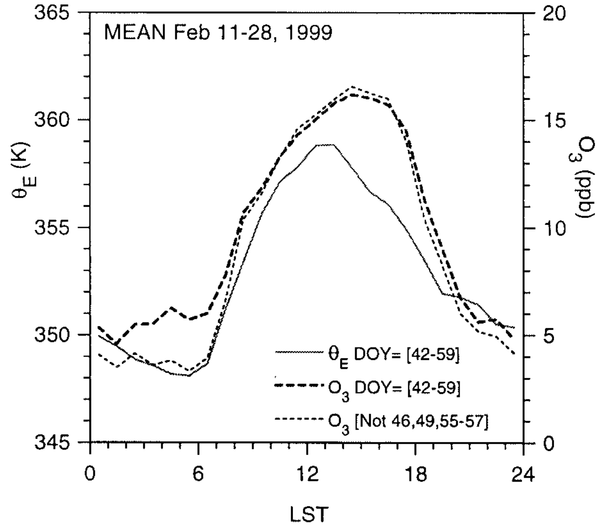


**Figure 1.** Mean diurnal cycle of $\theta_E$ and $O_3$ at surface from 11 to 28 February 1999.

### 2.1. Mean Diurnal Cycles of $O_3$ and $\theta_E$

[6] Figure 1 shows the mean diurnal cycles of surface $\theta_E$ (left-hand scale) and $O_3$ (right-hand scale) for the 19-day period from 11–28 February 1999 (days of year (DOYs) 42–59). The time axis is local standard time (LST), which is UTC- 4 hours. Because the surface cools at night, $\theta_E$ has a morning minimum at sunrise. The nocturnal stable BL largely uncouples the surface from the atmosphere above, so surface $O_3$ is generally low at night, because it is quickly removed by catalytic reactions. Two ozone averages are shown, one for the whole period and a second one (dotted), which excludes 5 days (DOYs = 46, 49, 55–57) when downdrafts bring down air at night from higher in the atmosphere (four of these cases are discussed in section 2.3). These two $O_3$ averages differ significantly only at night, when this second curve, influenced less by downdrafts, is lower and reaches 3 ppb just before sunrise. After sunrise, $\theta_E$ rises quickly, reaching a peak near 360 K just after local noon under the influence of the surface sensible and latent heat fluxes, themselves driven by the surface net radiation. As the surface $\theta_E$ rises, the BL deepens and mixes air down from higher in the atmosphere, which has a higher $O_3$ concentration. In addition, photochemical processes generate $O_3$ during the daytime. Thus at the surface, the morning rise of $\theta_E$ is closely coupled to a rise of $O_3$. In the afternoon, $\theta_E$ falls under the influence of precipitation-driven convective downdrafts, while $O_3$ stays high, reaching a mean value of 16 ppb at 1430 LST, because the same downdrafts that mix down lower $\theta_E$ air from higher in the atmosphere bring down air with probably higher $O_3$. As the surface cools and uncouples again from the atmosphere above in the late afternoon, $O_3$ again falls as it is removed by catalytic reactions.

### 2.2. Diurnal Cycle of Ozone Mixing Ratio During February 1999

[7] Figure 2 shows the diurnal cycle of ozone mixing ratio from 11 February to the end of February (DOYs 42–59). During the daytime hours, ozone concentrations are much higher during the period 11–21 February. This corresponds to a regime of northeasterly winds at low levels [*Rutledge et al.*, 2000; *Rickenbach et al.* 2002; *Betts et al.*, 2002]. At the end of Feb-

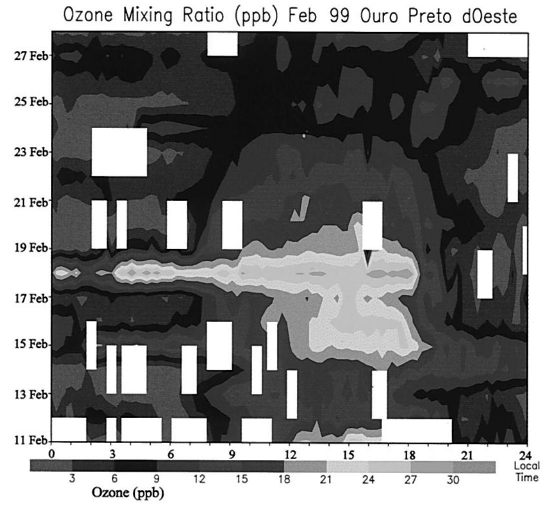


**Figure 2.** Ozone distributions in Rondônia from 11 to 28 February 1999. See color version of this figure at back of this issue.

ruary, when the winds have a westerly component and the convective regime is more maritime in nature (lower cloud tops, less lightning activity, and lower CCN (cloud condensation nuclei) counts [see *Rutledge et al.*, 2000; *Cifelli et al.*, 2002], daytime ozone concentrations are lower. Although nighttime ozone concentrations are generally quite low compared to those seen during the daytime, the night of 17–18 February, when a major convective squall-line passed the site, stands out as an exception, and will be discussed in more detail later. Other episodes of higher nighttime ozone can also be seen, particularly about 0200 LST on 15 February, events on 24 February in both morning and evening, and finally on 25 and 26 February in the evening. Each of them corresponds to occasions when convective downdrafts penetrate to the surface (see section 2.3).

### 2.3. Transport of $O_3$ and $\theta_E$ at Night by Convective Downdrafts

[8] Since surface $O_3$ is generally low at night, downdrafts at night, which couple the surface to the lower troposphere and bring down air with high $O_3$, are more readily visible at night than in the afternoon. Figure 3 has four panels, which show examples of this coupled vertical transport at night in the 18-day period. The top left panel shows the night of 17–18 February, when one of the strongest convective events of the month, a large squall line, passed over the site. The fall of $\theta_E$ and rise of $O_3$ are tightly coupled but exactly out of phase, as bursts of downdraft air reach the surface around 0000 and 0300 LST, indicating that the same vertical transport process is responsible for both changes. Surface $O_3$ reaches a maximum of 30 ppb, far above typical nocturnal values of 3–5 ppb and much higher than the afternoon mean of 16 ppb, shown in Figure 1. These high ozone values continue throughout the

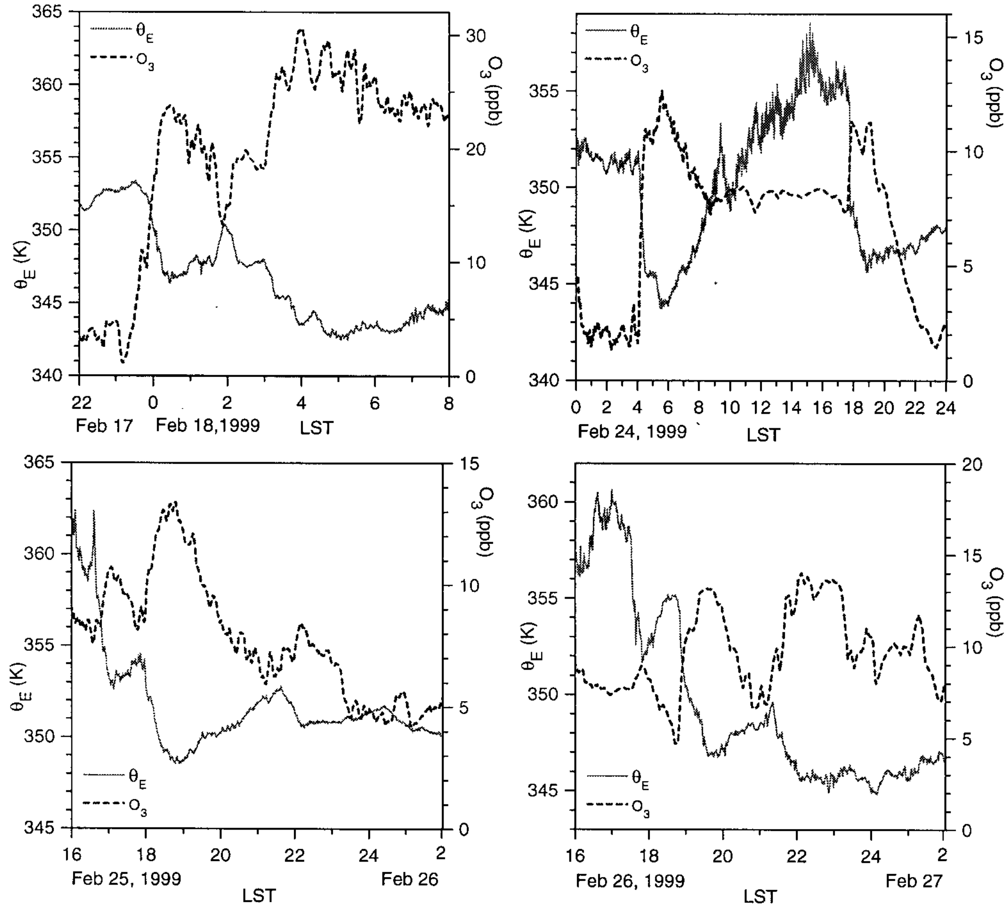


**Figure 3.** Coupling of high $O_3$ and low $\theta_E$ convective downdraft events at night.

following day, as shown in Figure 2. The other three panels show similar downdraft events on three other nights about a week later, which are not so extreme ($O_3$ rises to only ~12–14 ppb). The top right panel on 24 February is interesting as ozone peaks both before sunrise (0400–0500 LST) and near sunset (1800 LST), reaching concentrations that are higher than during the daytime period. The sudden drop of $\theta_E$ correlated with both these high-ozone episodes shows that the cause is downdraft air reaching the surface. The bottom two panels show further examples (seen earlier in Figure 2) near and after sunset, where the fall of $\theta_E$ caused by downdrafts is tightly coupled to a $O_3$ rise. The ozone traces show about a dozen similar large events at night during March and April 1999. Figure 2 also shows some weaker events at night (late on 11 February, early on 15 February, and early on 22 February), which are also associated with low $\theta_E$ downdrafts.

[9] Although we show only four events at night from this 18-day period, this does not mean that these downdraft events are infrequent. The rapid surface fall of $\theta_E$ associated with downdrafts can be seen with the passage of almost every convective band. However, the majority of the deep convective systems develop in the afternoon, when $\theta_E$ values and $O_3$ concentrations are high. For these afternoon events, the fall of $\theta_E$ is readily visible, as this transition typically only takes 30 min [*Betts et al.*, 2002], but the injection of midtropospheric high $O_3$ air can only be deduced from more detailed analyses [*Sigler et al.*, 2001].

### 2.4. Atmospheric Profiles of $\theta_E$ Near Downdraft Times

[10] Vertical profiles of $\theta_E$ from rawinsonde soundings launched at the pasture site near the downdraft times enable us to estimate roughly the probable level of origin of the downdraft air. Figure 4 shows four $\theta_E$ profiles, labeled with the time of launch, either before or close to the times of a downdraft event in each of the four panels in Figure 3. They all show with considerable fluctuations the general characteristic decrease of $\theta_E$ with height, although they have all been modified (cooled) between the surface and the 950 hPa. On each sounding we have circled the first level above the surface, where the $\theta_E$ reaches the low value seen in the downdraft air at the surface in Figure 3. If air descends in downdrafts with little mixing, and thus conserving $\theta_E$, this gives an estimate of the level of origin. The levels marked range from 765 to 874 hPa. Such originating levels for downdrafts are consistent with *Betts* [1976], who found that convective downdrafts over Venezuela generally came from the layer above cloud base. In Rondônia, afternoon cloud base was typically 100 hPa above the surface in the rainy season [*Betts et al.*, 2002]. Two of the rawinsonde profiles shown (on 17 and 24 February) were launched 1–2 hours before the surface passage of the convective downdraft, while the other two were launched during the downdraft event. Since we have no measurements of $O_3$ above the surface, we can say little about ozone chemistry within the deep convective systems. However, it is clear that the downward transport of ozone is rapid. The timescale of vertical advection within cumulonimbus from 800 hPa (given downdraft speeds of the order of meters per second) is only 10–20 min, much less than the lifetime of ozone in the free troposphere, usually estimated to be of the order of 14 days. Each rapid fall of $\theta_E$ and rise of $O_3$ at the surface, shown in Figure 3, also typically lasts ~20–30 min, with peak surface gusts reaching 6 m $s^{-1}$ (not shown).

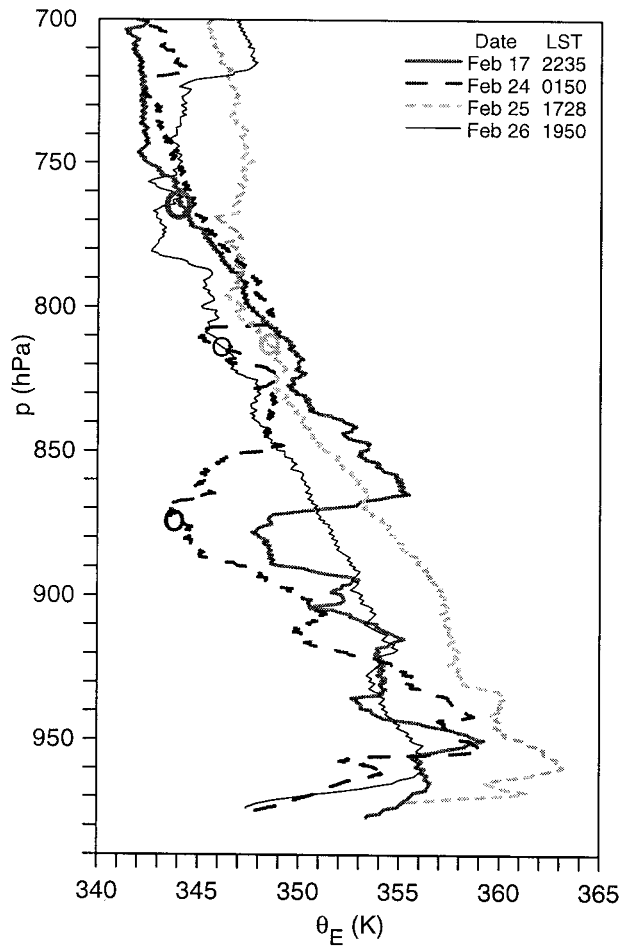


**Figure 4.** Soundings of $\theta_E$ near downdraft times.

### 2.5. 17–18 February Case

[11] Since the 17–18 February case was clearly exceptional, we present a satellite overview. Figure 5 shows a sequence of four satellite images, at 0345 UTC (top left), 1545 UTC (top right), 2145 UTC (bottom left) on 17 February, and bottom right, 0345 UTC on 18 February (2345 LST on 17 February), just as the leading convective system reaches the pasture site (marked as a red dot on these panels). This squall line had a very large-scale structure and time continuity, originating the night before near the northeast coast of Brazil, and propagating in a southwesterly direction till it passed the Rondônia region (indicated by the red box). This squall line is similar to the ones described previously by *Greco et al.* [1990], *Silva Dias and Ferreira* [1992], and *Cohen et al.* [1995]. The main feature of this type of system is that it begins with a few convective cells forced by the sea breeze circulation and may develop into a long-lived propagating squall line when there is a low-level (around 700 hPa) easterly jet in the vicinity. The particular system that reached Rondônia early in the morning of 18 February actually interacted with the remains of another convective system that was active late in the afternoon of 17 February, east of Rondônia.

## 3. Conclusions

[12] The tropical atmosphere is characterized by a decrease with height of equivalent potential temperature, $\theta_E$, because

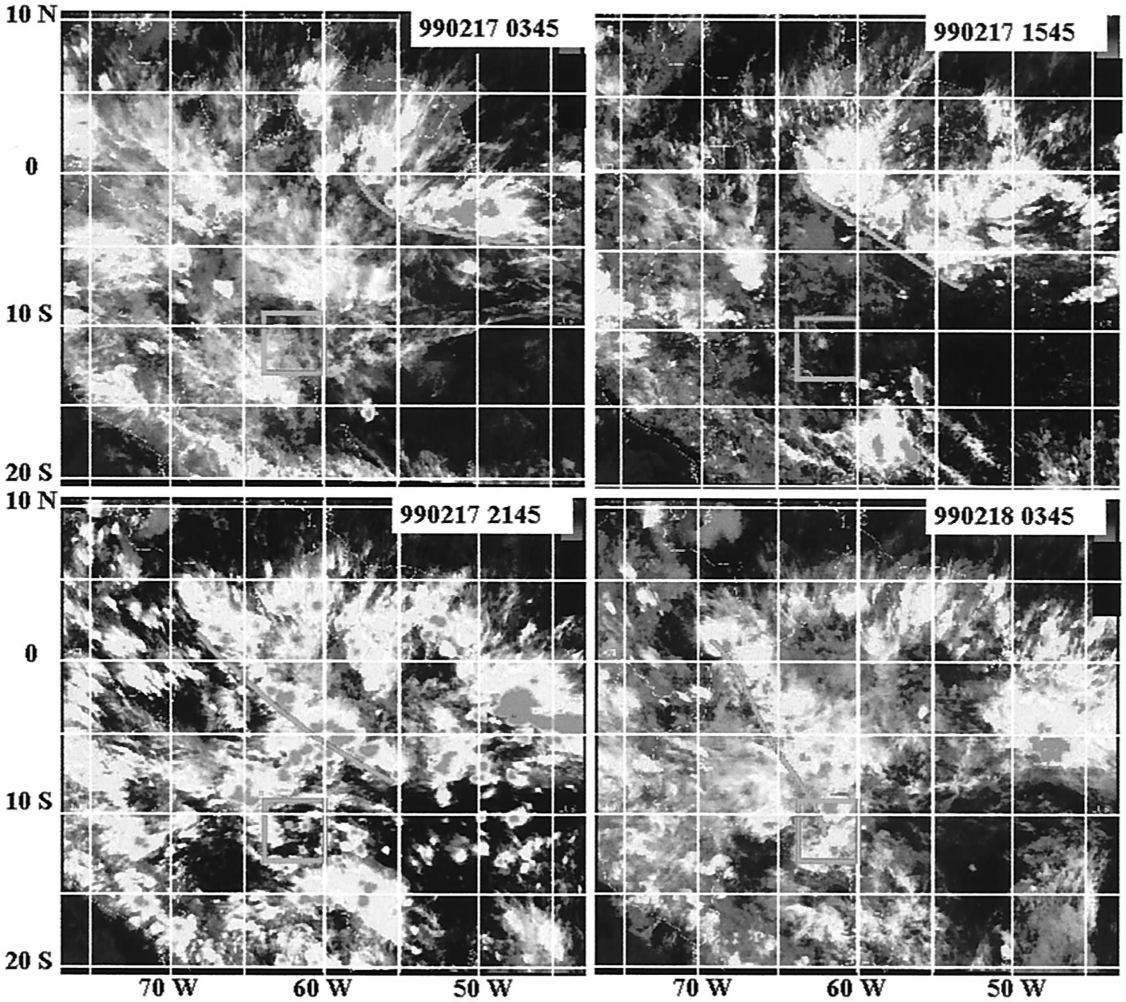


**Figure 5.** Sequence of satellite pictures, showing passage of squall line from the coast to Rondônia: top, 0345 and 1545 UTC on 17 February; bottom, 2145 UTC on 17 February, and 0345 UTC on 18 February. See color version of this figure at back of this issue.

the primary source of $\theta_E$ is from the daytime fluxes at the surface, and the primary sink is radiative cooling of the troposphere. Studies have shown that $O_3$ concentrations increase with height in the rainy season over Amazonia, because the forest is predominantly a sink of $O_3$ at that time. During the daytime, when vertical mixing is strong in the BL and there is some photochemical generation, surface $O_3$ values are high (~20 ppb). As the surface cools at night and the stable BL largely uncouples the surface from the troposphere above, near-surface $O_3$ values become low (~3–5 ppb) as $O_3$ is efficiently removed by surface deposition processes. Our measurements of $O_3$ and $\theta_E$ clearly show that nighttime convective downdrafts, driven by evaporation of falling precipitation, transport air with higher $O_3$ and lower $\theta_E$ rapidly down to the surface from the lower-middle troposphere, typically around 800 hPa. Although we show only four events at night from this 18-day measurement period, more detailed analyses [*Sigler*, 2001] for longer time periods show that this vertical $O_3$ transport is a pervasive process and can occur during night and day, whenever deep convective systems develop. Because the development of deep convective systems are almost a daily occurrence during the wet season [*Betts et al.*, 2002], our findings have important implications for the $O_3$ budget in the lower troposphere and its associated atmospheric chemistry. This downward transport of tropospheric ozone increases its deposition at the surface. During the wet season, higher boundary layer $O_3$ levels in the tropics can also impact two key processes. After undergoing photolysis, $O_3$ can be an important source for hydroxyl radicals. The unstable atomic oxygen resulting from the photolysis of $O_3$ can rapidly combine with water vapor to generate hydroxyl radicals, which are the most important chemical species in many photooxidation processes in the atmosphere. Finally, because $O_3$ is a highly phytotoxic and can be readily taken up by vegetation, increases in $O_3$ levels might reduce the net primary productivity of some vegetated ecosystems.

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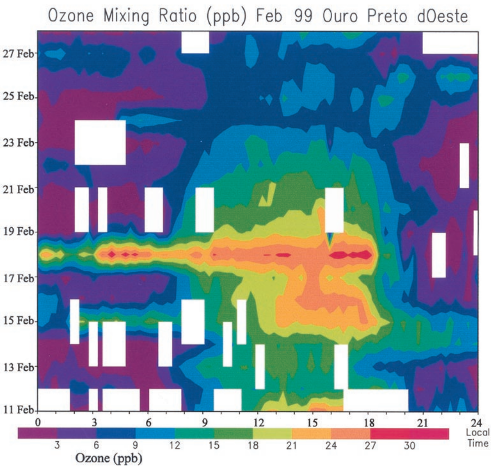
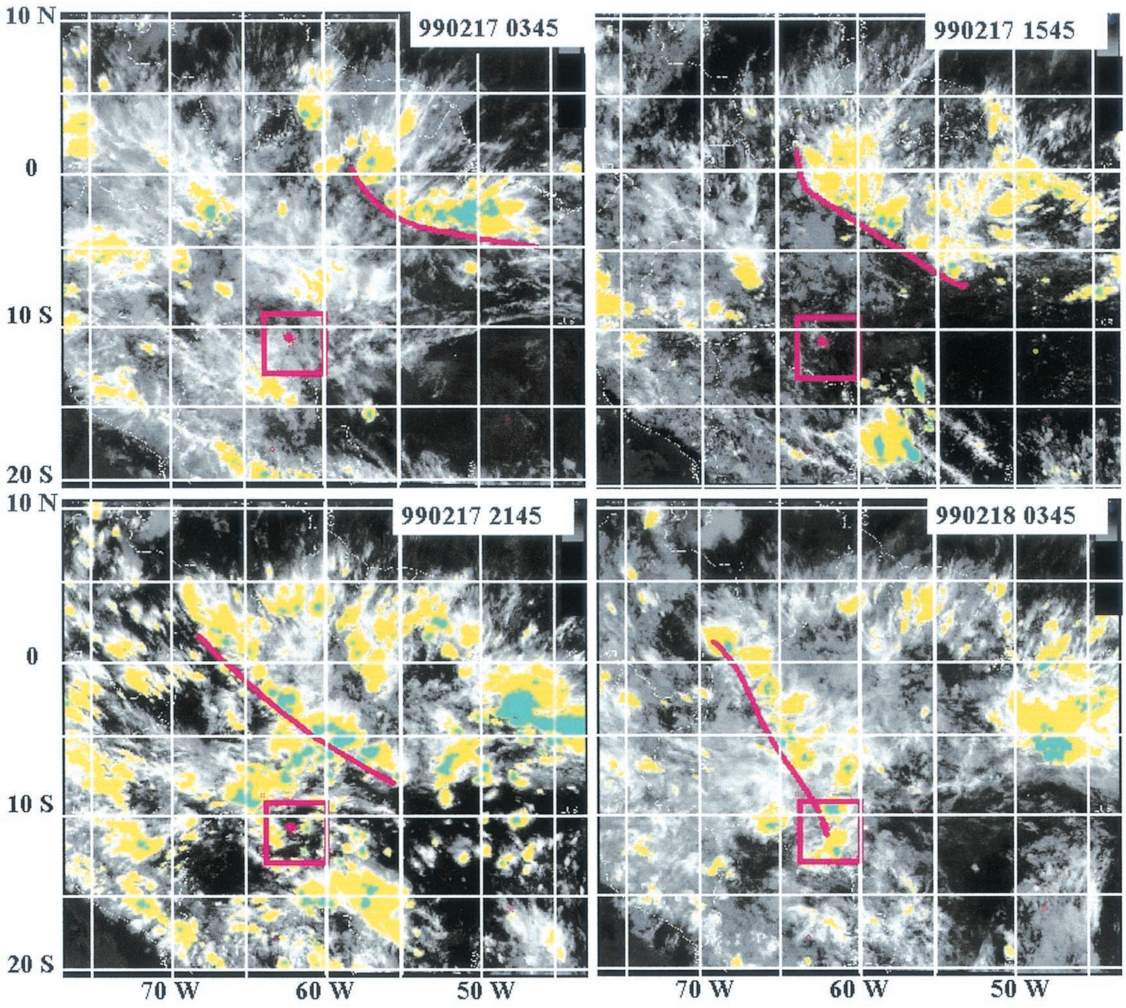



**Figure 2.** Ozone distributions in Rondônia from 11 to 28 February 1999.

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