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

Alan K. Betts[*]

Pittsford, VT, USA

Luciana V. Gatti,
Ana Maria Cordova

IPEN, Sao Paulo, Brazil

Maria A. F. Silva Dias

USP, Sao Paulo, Brazil

Jose D. Fuentes
Univiversity of Virginia, VA

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* Corresponding author:
Alan K. Betts, Atmospheric Research, Pittsford, VT 05763, USA
Email: akbetts@aol.com

ABSTRACT.

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 transport air with higher ozone and lower equivalent potential temperature down to the surface from around 800 hPa.

1. Introduction

The Wet season Atmospheric Mesoscale Campaign (WETAMC) of the Large-scale Biosphere-Atmosphere Experiment in Amazonia (LBA) in January and February of 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^o 46.42'S, 62^o20.22'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 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, non-pristine site, with a well-traveled highway within ten kilometers to the northeast. The landscape surrounding the measurement site is reasonably flat and situated 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 night-time data to show that convective downdrafts, which bring down air of lower equivalent potential temperature to the surface from the lower-middle troposphere, also bring down air of higher ozone concentration.

Equivalent potential temperature (θ_E) has long been used as a tracer for moist convective processes and transports, as it is conserved in the condensation and evaporation of water (e.g. Emanuel, 1994). The primary source of θ_E is at the surface, where the surface sensible and latent heat fluxes, driven by solar heating, can be considered a θ_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 θ_E with height. The tropical atmosphere is always close thermally to moist neutrality (e.g. Riehl, 1979; Betts, 1998), so that 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 θ_E (and so transport high θ_E air upward from the near surface mixed layer), while the evaporation of falling rain into unsaturated air also conserves θ_E to good approximation, and the cooled air sinks in downdrafts, which bring low θ_E air down from the lower-middle troposphere into the boundary layer (BL), and to the surface. Many papers have used θ_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 θ_E from one-min average micrometeorological measurements of pressure, temperature and humidity on a tower at a height of 1.5m (Betts et al., 2000) at this pasture site, and from radiosondes launched from the site.

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 largely to anthropogenic activity, could accumulate in the lower atmosphere to harmful levels. Conversely, tropospheric elevated ozone levels can contribute to altering the Earth's surface energy balance as ozone strongly absorbs thermal energy in the so-called atmospheric window (in the wavelength of ~ 9.6 µm). Free tropospheric values of ozone are high, as it is produced photochemically in the stratosphere, advected downward into the troposphere and eventually down to the ground surface. Near the surface in the atmospheric boundary layer, it can be both generated and removed by catalytic and photochemical reactions involving CO 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 stratospheric transport to the ozone levels recorded close to the ground. In the tropics, because processes such as tropopause folding occur at higher altitudes than in mid-latitude regions, 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). In the rainy season, ozone levels in Amazonia are reduced because of the suppressed photochemical activity resulting from cloudy conditions (and hence less actinic irradiance to drive ozone photochemistry) and 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., 1991; Sigler et al., 2000). Consequently, in the rainy season the Amazonian region is predominantly a sink of ozone, and ozone concentrations increase with height (Gregory at 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. As the surface cools and the stable BL largely uncouples the surface at night from the troposphere above, near-surface values become low because of dry deposition of ozone to the underlying surface (Kirchhoff, 1988; Fan et al., 1991). As a result, ozone levels immediately after sunset progressively decline, and the nocturnal BL becomes low in ozone due to surface deposition. However, in the presence of convective downdrafts, driven by the evaporation of falling precipitation, ozone levels can increase at night due to the strong coupling between the surface and 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. Relation between ozone and equivalent potential temperature at the surface

The coupling of ozone (O_3) and equivalent potential temperature (θ_E) is different during the daytime period, as 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.

2.1 Mean diurnal cycles of O_3 and θ_E

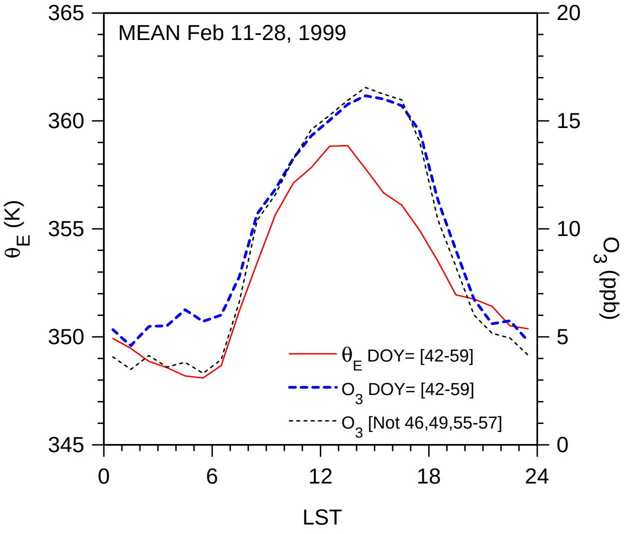

Figure 1. Mean diurnal cycle of θ_E and O_3.

Figure 1 shows the mean diurnal cycles of surface θ_E (left-hand-scale) and O_3 (right-hand-scale) for the 19-day period from February 11-28, 1999 [Day of year (DOY) 42-59]. The time axis is local standard time (LST), which is UTC- 4hrs. Because the surface cools at night, θ_E has a morning minimum at sunrise. The nocturnal stable BL largely uncouples the surface from the atmosphere above, so that surface O_3 is 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 (DOY = 46, 49, 55-57) when downdrafts bring down air from higher in the atmosphere at night (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 3ppb just before sunrise. After sunrise, θ_E rises quickly, reaching a peak just after local noon near 360K under the influence of the surface sensible and latent heat fluxes, themselves driven by the surface net radiation. As the surface θ_E rises, the BL deepens and mixes down air 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 θ_E is closely coupled to a rise of O_3. In the afternoon, θ_E falls under the influence of precipitation driven convective downdrafts, while O_3 stays high, reaching 16 ppb at 1430 LST, because the same downdrafts which bring down lower θ_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.

Figure 2 shows the diurnal cycle of ozone mixing ratio from the 11th to the end of February (DOY 42-59). During the daytime hours, ozone concentrations are much higher during the period February 11-21. This corresponds to a regime of north-easterly winds at low levels (Rutledge et al., 2000; Betts et al., 2000). At the end of February, 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), daytime ozone concentrations are lower. Although night-time ozone concentrations are generally quite low compared to those seen during the daytime, the night of February 17-18, 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 night-time ozone can also be seen, particularly about 2am LST on February 15, events on the 24th in both morning and evening, and finally on February 25 and 26 in the evening. Each of them corresponds to occasions when convective downdrafts penetrate to the surface (see next section).

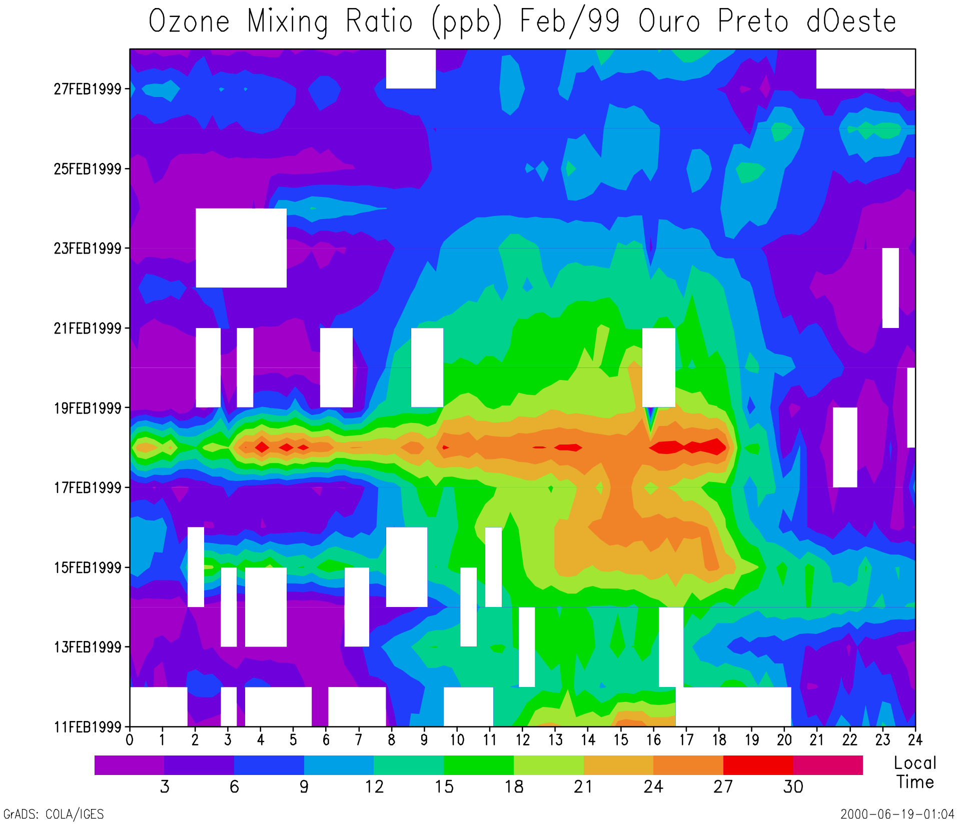

Figure 2 Ozone distributions in Rondonia from February 11-28, 1999.

2.3 Transport of O_3 and θ_E at night by convective downdrafts

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 4 panels, which show examples of this coupled vertical transport at night in the 18-day period. The upper left panel for the night of February 17-18 was one of the strongest convective events of the month, when a large squall-line has passed over the site. The fall of θ_E and rise of O_3 (note that the right-hand ozone scale has been *reversed* on Figure 3 from Figure 1) are closely coupled, as bursts of downdraft air reach the surface around 0 and 3 LST, indicating that it is the same vertical transport process responsible for both. Surface O_3 reaches a maximum of 30 ppb, far above typical nocturnal values of 5 ppb and much higher than the afternoon mean of 16 ppb, shown in Figure 1. These high ozone values continue throughout the following day, as shown on Figure 2. The other three panels show similar downdraft events on three other nights about a week later, which are not as extreme (O_3 rises to only about 12 ppb. The upper right panel on February 24 is interesting as ozone is higher both before sunrise (4-5 LST) and near sunset (18 LST) than during

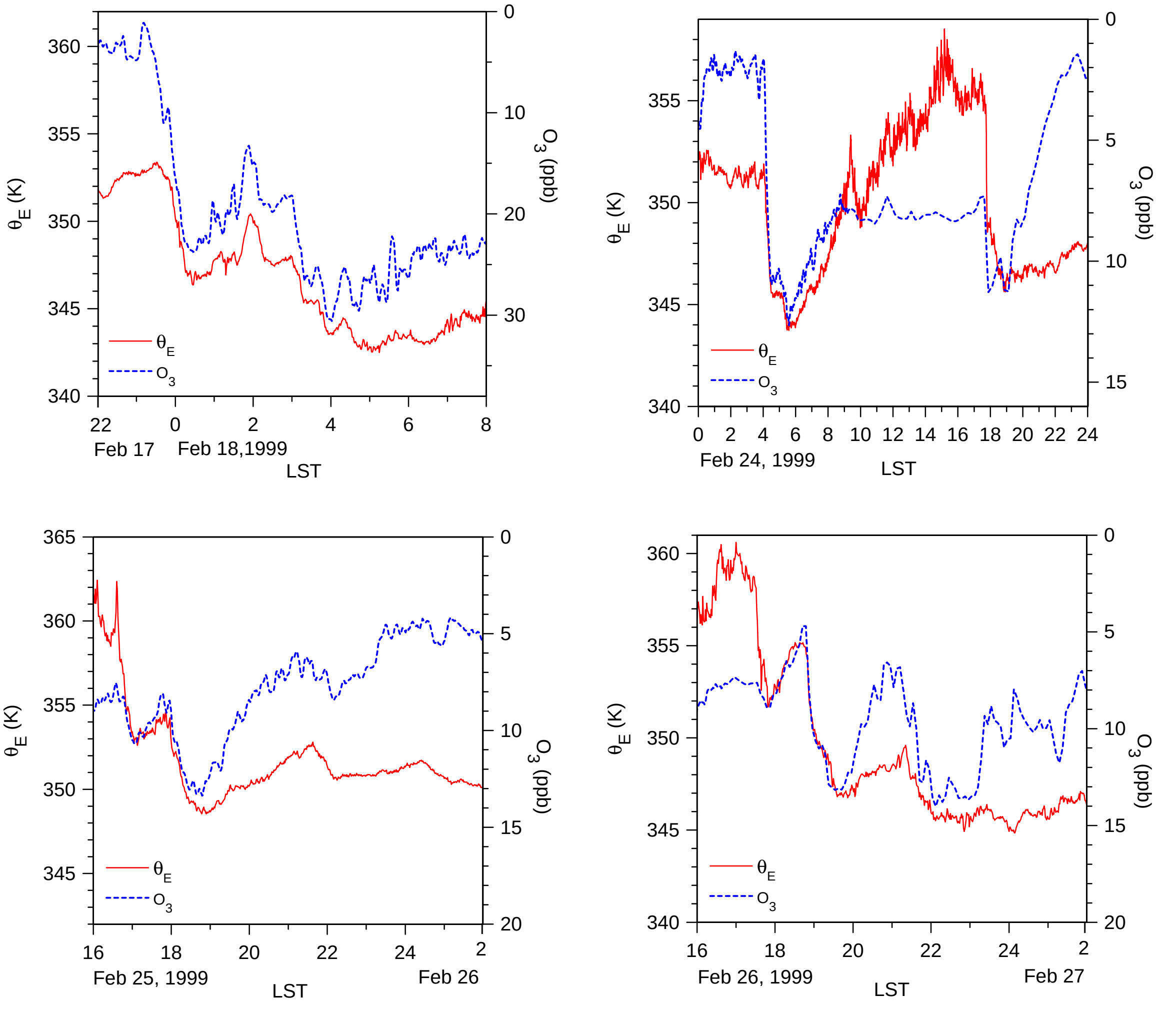

Figure 3. Coupling of high O_3 and low θ_E in convective downdraft events at night.

the daytime period. The sudden drop of θ_E correlated with both these high ozone episodes shows that the cause is downdraft air reaching the surface. The lower two panels show further examples (seen earlier on Figure 2) near and after sunset, where the fall of θ_E caused by downdrafts is tightly coupled to a O_3 rise. The ozone traces show more than a dozen similar large events at night during March and April, 1999. Figure 2 also shows some weaker events at night (late on the 11th, early on the 15th and early on the 22nd), which are also associated with low θ_E downdrafts.

2.4 Atmospheric profiles of θ_E near downdraft times

Vertical profiles of θ_E from soundings launched at the pasture site near the downdraft times enable us to estimate roughly the likely level of origin of the downdraft air. Figure 4 shows four θ_E profiles, corresponding to the four panels in Figure 3. They all show with considerable fluctuations the general characteristic decrease of θ_E with height, although they have all been modified (cooled) between the surface and 950 hPa. On each sounding we have circled the first level above the surface, where the θ_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 θ_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 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., 2000).

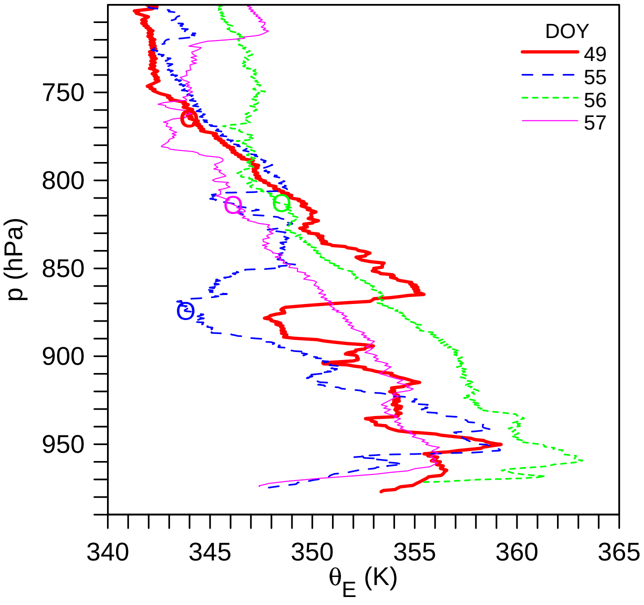

Figure 4. Soundings of θ_E near downdraft times.

2.5 February 17-18 case.

Since the February 17-18 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 the 17th Feb and at 0345 UTC on the 18th (2345 LST on the 17th) in the lower right, just as the leading convective system reaches the pasture site (see Figure 3, upper left panel). This squall-line had a very large scale structure and time continuity, originating the night before near the north-east coast of Brazil, and propagating in a south-westerly 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 the 18th February actually interacted with the remains of another convective system that was active late in the afternoon of the 17th , east of Rondônia.

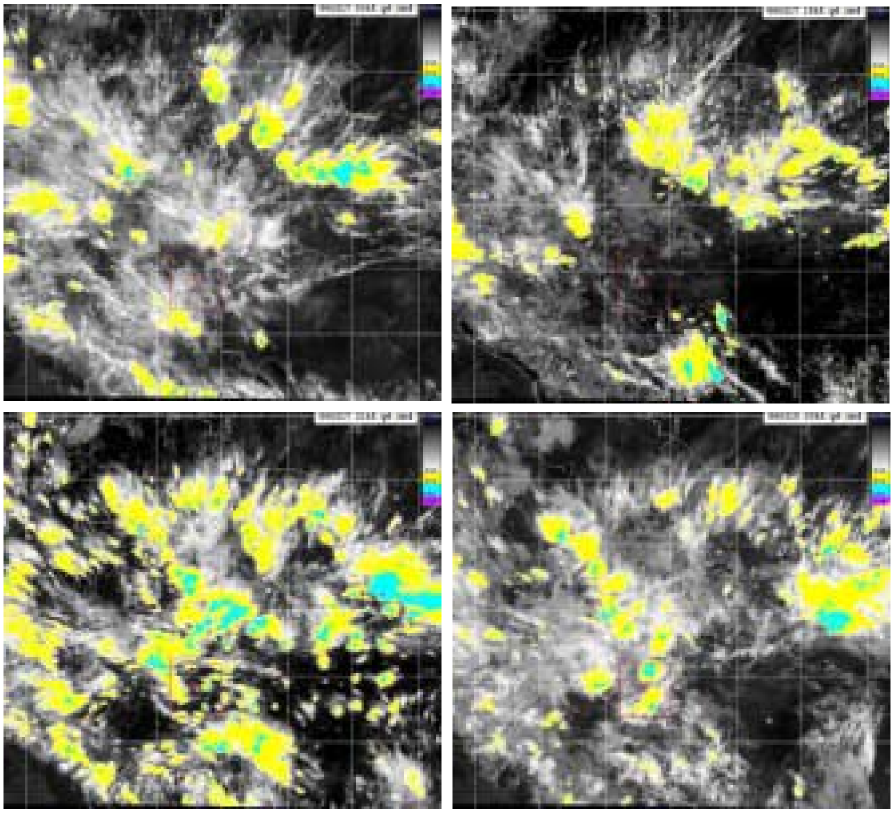

Figure 5 Sequence of satellite pictures, showing passage of squall-line from the coast to Rondonia: top, 0345 and 1545 UTC on 17th; bottom, 2145 UTC on the 17th and 0345 UTC on the 18th February.

3. Conclusions

The tropical atmosphere is characterized by a decrease of equivalent potential temperature, $\theta_{E,}$ with height, because the primary source of θ_E is at the daytime fluxes at the surface, and the primary sink is radiative cooling of the troposphere. Studies have shown that ozone concentrations increase with height in the rainy season over Amazonia, because the forest is predominantly a sink of ozone at that time. During the daytime, when vertical mixing is strong in the BL, surface ozone values are high, but as the surface cools and the stable BL largely uncouples the surface from the troposphere above at night, near-surface values at night become low, as ozone is removed by catalytic processes. Our measurements of O_3 and θ_E clearly show, when background levels of O_3 are low at night, that convective downdrafts, driven by evaporation of falling precipitation, transport air with higher O_3 and lower θ_E down to the surface from the lower-middle troposphere, typically around 800 hPa..

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