Convective storms and non-classical low-level jets during high ozone level episodes in the Amazon region: an ARM/GOAMAZON case study

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Abstract

In this work, we investigate the ozone dynamics during the occurrence of both downdrafts associated with mesoscale convective storms and non-classical low-level jets. Extensive data sets, comprised of air chemistry and meteorological observations made in the Amazon region of Brazil over the course of 2014–15, are analyzed to address several questions. A first objective is to investigate the atmospheric thermodynamic and dynamic conditions associated with storm-generated ozone enhancements in the Amazon region. A second objective is to determine the magnitude and the frequency of ground-level ozone enhancements related to low-level jets. Ozone enhancements are analyzed as a function of wind shear, low-level jet maximum wind speed, and altitude of jet core. Strong and sudden increases in ozone levels are associated with simultaneous changes in variables such as horizontal wind speed, convective available potential energy, turbulence intensity and vertical velocity skewness. Rapid increases in vertical velocity skewness give support to the hypothesis that the ozone enhancements are directly related to downdrafts. Low-level jets associated with advancing density currents are often present during and after storm downdrafts that transport ozone-enriched air from aloft to the surface.

Keywords: Ozone, storms, low-level jets, storm downdrafts, air chemistry, Amazon region.

1. Introduction

Ozone (O_3) is a greenhouse gas and knowledge of its temporal and spatial distribution can help determine its contribution to the Earth's surface energy balance (Mitchell, 1989). In the rural tropical atmospheric boundary layer (ABL), O_3 influences numerous chemical cycles (Wofsy and McElroy, 1974). For example, O_3 reacts with soil-emitted nitric oxide (NO) to form nitrogen dioxide (NO_2) and molecular oxygen (O_2), *i.e.*, $O_3 + NO \rightarrow NO_2 + O_2$. The photolysis of NO_2 generates NO and atomic oxygen (O) which readily combines with O_2 to reform O_3 in the presence of a third molecule such as nitrogen (N_2). Also, O_3 can undergo photolysis to generate O_2 and electronically excited oxygen atoms ($O(^1D)$), that is, $O_3 + h_\nu \rightarrow O(^1D) + O_2$. In rich water vapor (H_2O) environments such as over the Amazon rainforest, the $O(^1D)$ rapidly combines with H_2O to produce hydroxyl radicals (OH) (Finlayson-Pitts and Pitts, 2000), *i.e.*, $O(^1D) + H_2O \rightarrow OH + OH$. In addition, O_3 reacts with rainforest-emitted hydrocarbons to produce high yields of OH (Gerken et al., 2016) and reaction products that can condense and form secondary organic aerosols. OH is considered the *cleanser* of the atmosphere and as such can act as the main sink of gases such as methane (CH_4) (Hu et al., 2010a). Knowledge of OH sources and sinks is necessary to determine the oxidation capacity of the atmosphere. Ozone can also be removed from the atmosphere via surface deposition (Sigler et al., 2002).

In the tropical troposphere O_3 levels increase with altitude in response to the transport of O_3 -rich air from the lower stratosphere to the upper troposphere (Garstang et al., 1988; Kirchhoff et al., 1990). Immediately above the rainforest O_3 varies rapidly with altitude due to the dominance of sink processes such as surface deposition and chemical reactions that result in O_3 mixing ratios approaching 0 parts per billion on a per volume basis (ppbv) near the forest canopy. In the tropics, during the wet season, deep mesoscale convective storms redistribute trace gases and aerosols throughout the troposphere (Garstang et al., 1988; Scala et al., 1990). In the updraft region of storms, boundary layer air is carried to the upper troposphere whereas within the storm downdraft air parcels are transported from the middle of the troposphere to the surface. Such circulation patterns within storms generate unique chemical signatures in the ABL. For instance, in the areas impacted by the downdraft region of convective storms,

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66 ground-level O_3 can suddenly increase in response to the transport of air parcels
 67 from the middle of the troposphere where O_3 levels are considerably higher than
 68 at the surface (Kirchhoff et al., 1990). Ground-level O_3 enhancements caused by
 69 storms depend on several factors, including intensity of atmospheric convection,
 70 storm propagation speed, origin of downward moving air parcels, and time of
 71 the day (Betts, 2002; Sahu and Lal, 2006; Grant et al., 2008; Hu et al., 2010b;
 72 Gerken et al., 2016). The O_3 enhancements can be as much as 30 ppbv.

73 With the exception of the study by Gerken et al. (2016) that included almost
 74 one year of field observations over the rainforest of Brazil, most studies report
 75 storm-related O_3 enhancements for brief field campaigns.

76 The processes governing O_3 enhancements in the atmospheric boundary layer
 77 over the rainforest, associated with storms, are crucial to understand the chem-
 78 ical cycles involved with the formation of oxidations and aerosols. For exam-
 79 ple, the Amazon rainforest emits large amounts of isoprene, monoterpenes, and
 80 sesquiterpenes (Guenther, 2006; Jardine et al., 2015). Such gases react with O_3
 81 to form free radicals (e.g., OH) and precursors of secondary organic aerosols
 82 (Fuentes et al., 2000). Therefore, one objective of the present study is to investi-
 83 gate the atmospheric thermodynamic (e.g., convective available potential energy
 84 (CAPE)) and dynamic (e.g., standard deviation (σ_w) and skewness (Sk_w) of the
 85 vertical velocity) conditions associated with storm-generated O_3 enhancements
 86 in the Amazon rainforest. A second objective is to determine the magnitude
 87 and the frequency of ground-level O_3 enhancements related to the appearance of
 88 “non-classical” low-level jets (LLJ’s) — in the sense that they are different from
 89 classical nocturnal LLJ’s (see for example Banta et al., 2006) — that develop
 90 in response to those storms. For this purpose, air chemistry and meteorological
 91 data sets, obtained as part of an unprecedented year-long measurement campaign
 92 in the Amazon region within the context of the GoAmazon project (Martin et al.,
 93 2015), are analyzed and interpreted, and the LLJ’s associated with O_3 enhance-
 94 ments are identified with advancing density currents originating from the storm
 95 cold downdrafts.

96 **2. Methods**

97 *2.1. Experimental Site*

98 The data used in this study were obtained at the rural site near Manacapuru
 99 (3.2°S, 60.6°W, 34 m above sea level, also known as T3) which is located at ap-
 100 proximately 70 km from the City of Manaus, Amazonas, Brazil (Fig. 1). Grass
 101 covers the land around the measurement station and beyond the study site farm-
 102 land is the dominant land use. Rainforest dominates the regional landscape. Data

Figure 1: The map indicates the location of the study site with reference to the City of Manaus, Amazonia, Brazil. The photograph (taken by the DOE ARM team members) shows the surface characteristics of the instrumented study site.

sets were obtained as part of the U.S. Department of Energy Atmospheric Radiation Measurement Program (ARM, <http://www.arm.gov/measurements>) during March 2014 to September 2015. The deployment of the ARM facility took place during the GoAmazon 2014–15 project (Martin et al., 2015).

2.2. Air chemistry and meteorological measurements

Ozone mixing ratios were measured with an ultra violet light absorbed gas analyzer (model 49i, Thermo Fisher Scientific Inc., Waltham, MA, with an accuracy of 2 ppbv or 5%, whichever is greater). The instrument outputs a new measurement every 4 s, and was installed at a height of 3.5 m above the ground; it had in its inlets a 1- μ m pore size teflon membrane to keep the air sampling tubing clean from any dust or pollen. Other trace gasses (i.e., CO, NO, NO₂) were measured using similar setups to O₃. Meteorological data were collected at several heights above the ground near the aerosol observation site.

Upper air data were measured between October 2014 to June 2015. CAPE, defined as

$$\text{CAPE} = \int_{z_f}^{z_n} g \frac{T_{v,sa} - T_v}{T_v} dz \quad (1)$$

was determined from microwave radiometer profiler (MWRP) measurements made at a sampling rate of 1 Hz. In Eq. (1), z_f is the level of free convection (LFC), and z_n is the equilibrium level (EL); $T_{v,sa}$ is the virtual temperature profile of a saturation adiabat starting at the lifting condensation level (LCL), and T_v is the observed virtual temperature profile (Iribarne and Godson, 1986). In this work, we used the CAPE data already calculated and available in the datasets described in sub-section 2.1.

The MWRP works with frequencies between 22–30 GHz and 51–59 GHz. Furthermore, the MWRP provides vertical profiles of temperature, humidity and cloud liquid water content as a function of height.

Profiles of σ_w and Sk_w were estimated from Doppler Lidar vertical velocity and cloud statistics (DLWSTATS). This device produces height- and time-resolved estimates of vertical velocity variance, skewness, and kurtosis every 1 s and with a vertical resolution of 30 m. Moreover, it also estimates cloud properties from the vertically pointing measurements, including cloud-base height, cloud frequency, cloud-base vertical velocity and cloud-base updraft fraction.

All the data were averaged into 5-, 10-, and 30-minute blocks. We used some meteorological variables such as barometric pressure, relative humidity and air temperature to calculate the equivalent potential temperature, θ_e . Events of sudden increase of surface O_3 were identified by an algorithm devised to detect O_3 concentration (mole fraction, in ppbv) variations (in time) ΔO_3 that had to satisfy the criteria of at least 3 ppbv in 5-min. averages and a simultaneous decrease in θ_e of 2.5 K within a 1-h time window of the event (Gerken et al., 2016). During these O_3 -enhancement events, a ΔCAPE corresponding to the maximum minus the minimum CAPE value during this one-hour window was also calculated.

We also used satellite and radar images in order to assess convective activity during the occurrence of O_3 -enhancement events. For this, NOAA GOES 13 images and an S-band (10-cm wavelength) Doppler radar with a 500-m gate spacing and a 1.8° beam width across the Amazon were employed. The S-band Doppler radar belongs to *Sistema de Proteção da Amazônia* (SIPAM) and is located at the Manaus airport ($3^\circ 08.98' \text{ S}$, $59^\circ 59.48' \text{ W}$, elevation 102.4 m). The operational scan strategy swept 17 elevation angles every 12 min and data were recorded for scans extending up to 250 km from the radar site.

Finally, 1-h sodar profiles were used for selecting low-level jet (LLJ) events occurring above the study site. The LLJ data analyzed here were measured from 25 March 2014 to 17 September 2015.

For the identification of low-level jets the following criteria were used (the j subscript indicates the height of the jet maximum): i) the first wind-speed maximum (U_j) above ground level has a speed of at least 1.5 m s^{-1} greater than the adjacent minima above and below (Banta et al., 2006; Duarte et al., 2015); ii) U_j must be greater than or equal to 4 m s^{-1} and occur below a height of 1000 m (Andreas et al., 2000; Santana et al., 2015). With this method, we extracted the relevant LLJ information, such as height (Z_j), local maximum (U_j), and wind direction (α_j) at the jet's core.

Both “classical” and “non-classical” LLJ's were identified with these criteria. The “non-classical” LLJ's are associated with the O_3 enhancement events at the surface, and can be identified with density currents, as will be shown in the next section.

3. Results and discussion

3.1. Case study of an ozone-enhancement event

During the 2014/15 GoAmazon-campaign, O_3 -enhancement events were detected during several occasions. To motivate some of the statistical analyses that will be performed, we begin by presenting one of these occasions (22 November 2014, Figure 2). On this day, there was a strong O_3 -enhancement (approximately 30 ppbv) around 0230 h UTC (at this site, UTC is equal to local time + 4 h). The data shown are, respectively, the time series of O_3 (Fig. 2-a), θ_e (Fig. 2-b), CAPE (Fig. 2-c), NO and NO_2 (Fig. 2-d), CO (Fig. 2-e), and finally horizontal wind speed (Fig. 2-f). This O_3 -enhancement event was associated with: i) a decrease of the θ_e value (approximately 11 K); ii) a decrease of the CAPE value (approximately 1600 J Kg^{-1}), iii) a decrease and increase of NO and NO_2 levels, respectively, iv) a decrease of CO concentration (to approximately 40% of its initial value), and v) a strong increase of horizontal wind speeds near the ground. This is a pattern that, by and large, is repeated for almost all O_3 -enhancement events. Its phenomenology is now largely accepted to be related to storm downdrafts bringing cold and dry air (low θ_e), rich in O_3 and poor in CO, from upper layers to the surface, where O_3 reacts with soil-emitted NO to form NO_2 . Until now, however, this mechanism had been explained experimentally mostly on the basis of the θ_e surface observations.

More recently, however, Costantino and Heinrich (2014) have shown evidence, both from numerical simulations and data observations, that the propaga-

tion of density currents is associated with the cold-air downdrafts from tropical convective cells. The signature of the density currents on near-surface measurements are sudden changes in θ_e , air density, atmospheric pressure, wind speed and wind direction. Moreover, the thickness of the advancing cold air pool in Costantino and Heinrich’s simulations can exceed 1 km above the surface, and that is a region of increased wind speeds (*c.f.* their Figure 10). The wind speed decreases significantly above the cold air region. A considerable increase in near-surface horizontal wind speed has also been observed in other O₃ and convective storm studies (Grant et al., 2008; Wissmeier and Goler, 2009; Rafkin and Barth, 2015).

Figures 2-b and 2-f show the same pattern identified by Costantino and Heinrich (2014) with an advancing density current (for θ_e and wind speed). To present further evidence that a density current is passing by the measurement site, Figure 3 shows the corresponding traces of atmospheric pressure, air density and wind direction. Both air density and atmospheric pressure attain maxima at the time of the event. Wind direction fluctuates around 0/360°, but a small yet discernible change of direction is also seen at 0230 h UTC. The same pattern regarding density currents shown in Figures 2–3 was observed for all significant episodes of increased ozone ($\Delta\text{O}_3 > 8$ ppbv; see Section 3.5 and Figure 12). These increased O₃ events are associated with a high frequency of occurrence of low-level jets. The surface and upper-air data therefore indicate that these “non-classical” LLJ’s during the episodes of ozone increase are actually the signature of the density currents across the disturbed atmospheric boundary layer. Finally, Figure 4 confirms the presence of a mesoscale convective complex above the measurement site (indicated by a star) at the time of the event.

3.2. The ensemble behavior of CAPE during ozone-enhancement events

In Fig. 2-a and Fig. 2-c it was observed that during an O₃-enhancement event there was a decrease in the CAPE value. As shown by Chase et al. (2016), there is a relationship between CAPE before the storm and O₃-enhancement events. Figure 5 shows the ensemble behavior of CAPE for all the O₃-enhancement events detected throughout 2014–15. In the figure, CAPE is plotted from 1.5 h before to 2.0 h after the nominal time when ozone concentrations increased.

At 1.5 h before the events, CAPE is on average 1800 J kg⁻¹. It is steadily reduced up to the time when an increase in O₃ mixing ratio is detected, when it reaches an average of 500 J kg⁻¹. CAPE increases after this point, but does not recover the values before the event for a considerable time. Although the figure only shows the first two hours following the ozone increase, CAPE will typically only reach back to 1800 J kg⁻¹ a few hours after the event. This result confirms

Figure 2: Case study of a storm-caused ozone enhancement on 22 November 2014. Time series of (a) ozone mixing ratio, (b) equivalent potential temperature, (c) CAPE, (d) reactive nitrogen oxides, (e) carbon monoxide and (f) horizontal wind speed. The gray line represents the time at which ozone starts to increase

Figure 3: The signature of a density current passing by the T3 site: (a) air density; (b) atmospheric pressure and (c) wind direction.

Figure 4: NOAA GOES 13 Image for the Nov 2014, 0230 UTC O_3 -enhancement event

Figure 5: Changes in CAPE values 1.5-h before and 2.0-h after ozone enhancement events in 2014–15. The shaded area corresponds to one standard deviation around the average value.

that O_3 enhancement events occur after periods of elevated CAPE, which is a requirement for the occurrence of storms.

Because of the notable changes in CAPE depicted in Fig. 5, we show ΔO_3 against $\Delta CAPE$ in Fig. 6-a. Interestingly, changes in CAPE are better correlated to ΔO_3 than $\Delta \theta_e$, as can be seen in Fig. 6-b. Note that θ_e is the original marker used to identify the origin of the O_3 (Scala et al., 1990; Betts, 2002). Fig 6-a suggests that high/low ΔO_3 values are associated with high/low reductions in the CAPE values, but a caveat is in order. We found that not all situations of large reductions in CAPE values were associated with O_3 -enhancement events. In order to do that, we identified all situations where CAPE showed a reduction equal to or greater than 500 J kg^{-1} . This value was chosen on the basis of the regression line in Fig. 6-a, and it corresponds (on that figure) to $\Delta O_3 = 3 \text{ ppbv}$.

Figure 6: Relationship between ozone enhancements and decreases in: a) CAPE and b) equivalent potential temperature θ_e during the occurrences of storms in 2014.

For these situations, only 54% of the cases were associated with O_3 -enhancement events. In the remaining cases (46%), O_3 did not increase by at least 3 ppbv. In summary, for every O_3 -enhancement event there is a significant reduction of CAPE. The contrary is not true: not every reduction of CAPE is related to increases in O_3 at this site.

3.3. Are the ozone enhancement events related to the clustering of precipitation cells?

Recently, it has been suggested that different patterns of O_3 enhancement can be associated with different types of precipitation (Gerken et al., 2016). These authors identified “basin-wide” (the term meaning the scale of the Amazon region) precipitation events associated with increases of surface O_3 . The subjective analysis of precipitation fields in radar images, however, tends to limit the number of events analyzed.

To better understand the relationship between different types of convective activity (single cell or multiple cell) and possible different transport mechanisms of O_3 -rich air from the mid-troposphere to the Earth’s surface, CAPE and wind shear near the surface are jointly considered for each O_3 enhancement event.

CAPE and wind shear may explain the different behaviors of deep convection in the Earth atmosphere (Weisman and Klemp, 1982; Rotunno et al., 1988; Weisman and Rotunno, 2004; Wissmeier and Goler, 2009; Rafkin and Barth, 2015). The explaining variable is the bulk Richardson number (Wissmeier and Goler, 2009; Rafkin and Barth, 2015)

$$Ri_b = \frac{CAPE}{\frac{1}{2}u'^2} \quad (2)$$

where u' is the weighted wind shear between $z = 0$ and $z = 3$ km. Note that Wissmeier and Goler (2009) used a different range for the wind shear, $z = 0$ and $z = 6$ km. On the other hand, both Halverson et al. (2002) and French and Parker (2012) have found the layer between 0 and 3 km to be a better choice, since the low-level shear may be more important for large convective cells. During TRMM-LBA (Tropical Rainfall Measuring Mission – Large Scale Biosphere-Atmosphere Experiment in Amazonia) the layer between 0 and 3 Km seems to capture the most significant variation of wind structure in the lower and middle troposphere (Halverson et al., 2002). In the present work, the most significant vertical wind variation was also in this range.

According to Rafkin and Barth (2015), it is expected that high Ri_b values be associated with “single cell” events while low values may indicate the presence of “multiple cells”. Careful visual inspection of radar images confirms that this association between Ri_b and type of precipitation holds for the present data (i.e., lower/higher values of Ri_b were related to situations where the radar images showed large/small areas of precipitation).

It can be seen (Fig. 7) that there is no discernible relationship between ΔO_3 and Ri_b : the increases of O_3 levels are more or less evenly distributed among 3 orders of magnitude of Ri_b . Given that radar images confirm that Ri_b is a good predictor of the type of convective activity, in our data set the O_3 -enhancement events caused by “single cells” and “multiple cell” precipitation events are similar.

3.4. *Downdrafts and ozone-enhancement events*

The GoAmazon experiment provided a unique opportunity to study the upper-air characteristics of the atmosphere in connection with O_3 -enhancement events. Having considered upper-air characteristics from the point of view of CAPE and Ri_b , we now turn to the profiles of vertical velocity statistics.

In order to provide a baseline for comparison, three typical days (05, 08 and 14 November 2014) without precipitation, during which there no O_3 -enhancement

Figure 7: Relationship between ozone enhancement and the Richardson number (R_i) during the occurrences of storms in 2014

Figure 8: Vertical variation of: a–c) the standard deviation (σ_w) and d–f) skewness (Sk_w) of the vertical wind speed for three typical days (05, 08 and 14 November 2014) without ozone enhancements.

290 events, are shown in Figs. 8a–f. Figures 8 a–c show the profiles of σ_w (related to
 291 turbulence intensity) and Figs. 8 d–f show the profiles of Sk_w (positive and neg-
 292 ative values of Sk_w are related to strong updrafts and downdrafts, respectively).
 293 Overall, there is moderate turbulence intensity and a prevalence of stronger up-
 294 draft and weaker downdrafts in the afternoon periods.

295 In Fig. 9 we show the same data for three days (14 April, 28 May, and 13
 296 July) in which it was possible to observe large values of ΔO_3 at the surface. Ac-
 297 cording to the Ri_b criterion, two of those days (14 Apr and 13 July) correspond to
 298 “multiple cell” events, while the other one (28 May) corresponds to the presence
 299 of a “single cell”, as discussed earlier.

300 In situations when there are no O_3 -enhancement events (Fig. 8) it is noted
 301 that between 1200 h and 2100 h the turbulence intensity increased and Sk_w is
 302 predominantly positive. This situation is characteristic of convective ABLs and it

Figure 9: Vertical variation of: a–c) the standard deviation (σ_w) and d–f) skewness (Sk_w) of the vertical wind speed for three typical days (04-14, 05-28 and 07-13, 2014) with O_3 enhancements. The dotted red lines correspond to the period in which it was possible to observe the presence of downdrafts and O_3 -enhancement

Table 1: Values of ΔO_3 and $\Delta \theta_e$ determined for the episodes of convective rain shown in Figs. 9-a–f.

Date	ΔO_3 (ppbv)	$\Delta \theta_e$ (K)
04-14	17.9	11.4
05-28	18.0	3.4
07-13	10.8	10.1

Table 2: Statistics of the LLJ events (1-h profiles) observed from Mar 2014 to September 2015

Z_j (m)	570 ± 209
U_j (ms^{-1})	9.2 ± 2.9
U_j/Z_j (s^{-1})	0.018 ± 0.008
wind shear bellow LLJ core (s^{-1})	0.014 ± 0.009
Number of events (LLJ)	915
Number of profiles used in the analysis	9415

is expected in the absence of storm downdrafts. On the other hand, for situations where there are O_3 -enhancement events (Fig. 9), the turbulence intensity shows relatively high values and Sk_w is strongly negative, at around the same time when there is an O_3 -enhancement event.

Both “multiple cell” and “single cell” events have the ability to generate downdrafts, apparently with similar intensity ($\text{Sk}_w \approx -3$ in both cases). Figs. 9-a–f and Table 1 reveal that the O_3 enhancement events show relatively close values of O_3 for both “single cell” and “multiple cell” episodes.

Similar results were found by Grant et al. (2008), who analyzed data collected 40 km southeast of Dakar, Senegal. They noted that the arrival of a storm was responsible for the increased intensity of turbulence for about two hours. They reported that high levels of turbulence and high values of horizontal wind speed were strong enough to carry O_3 down toward the surface, increasing O_3 levels from 5–10 to 30 ppbv. Hu et al. (2010b) simulated convective storms in West Africa and showed that O_3 -enhancement events were directly impacted by the storm. According to Hu et al. (2010b), larger ΔO_3 values coincided with storm regions exhibiting maximum reflectivity values.

3.5. Low-level jets and ozone-enhancement events

The main features of the LLJ events observed in this study are included in Table 2. The total number of 1-h blocks associated with LLJ events was 915, and the number of 1-h sodar profiles used in the analysis was 9415 (i.e., $\sim 10\%$ of all wind profiles analyzed presented LLJ events). Our analysis shows an average of 51 LLJ events per month. September and October had a high incidence of LLJ events (30% of all cases). On the other hand, June and July had the lowest occurrences of LLJ events, with only 10%. Note that this analysis includes *all* LLJ’s observed at the site, and not only those related to O_3 -enhancement events.

Fig. 10 shows histograms of LLJ height, speed, and direction. For LLJ height, higher frequencies were observed in the 300 – 500 m range, especially

Figure 10: Frequency of occurrence of (a) maximum wind speed at the core of the low-level jet, (b) core of the low-level jet altitude, and (c) prevailing wind direction during low-level jet events in 2014.

at 500 m, while LLJ speed had a frequency peak in the 8 – 12 m s^{-1} range. Regarding jet direction, a higher frequency of occurrence was observed in the East quadrant. This direction is similar to the predominant wind direction at Manacapuru, that is, the direction of the Trade winds. The values of Z_j and U_j are close to those found by Santana et al. (2015) for data collected above Fazenda Nossa Senhora, a deforested area in the Amazon forest at the state of Rondônia, Brazil.

In order to illustrate the general shape of the LLJs above the Manacapuru site, Fig. 11-a shows the vertical variation of wind speed during the occurrence of a low-level jet at 02:00 h on 22 November 2014 (for this day, the time series of O_3 , θ_e , CAPE, NO, NO_2 , CO, and wind speed are shown in Fig. 2, and the occurrence of the LLJ is approximately coincident with the O_3 enhancement highlighted by the grey line). Fig. 11-b shows the ensemble of mean wind-speed profiles for

all cases analyzed in this study (black line with one standard-deviation bars) and also the ensemble of the mean wind-speed profile for all LLJs analyzed by Duarte et al. (2015) (gray line). The mean profiles of LLJs show considerable similarity, both above and below the LLJ core (Fig. 11-b). It is worth noting that these profiles are from regions with different topographies and climatologies. This similarity suggests a certain generality for the shape of the wind profile during LLJ events. As highlighted by Duarte et al. (2015), it can be important for advances in the numerical simulation of LLJs.

The above statistics relate to all LLJs observed, regardless of O_3 changes at the surface. From the opposite perspective, additional analysis of wind profiles during the O_3 -enhancement events revealed a high proportion of formation of LLJs. Although for 47% of the 166 O_3 -enhancement events identified in this study the wind profiler was not working, for the remaining 53% (88 cases), there were no LLJ events for situations with $\Delta O_3 \leq 8$ ppbv (38% of cases), as shown in Fig. 12. For all other cases, there were LLJ events present. Moreover, there is little relationship between ΔO_3 and the strength of LLJ (measured by the wind shear below of the core of the jet), see Fig. 13-a, despite the connection between larger decreases of $\Delta \theta_e$ and stronger LLJ's shown in Fig. 13-b. This is confirmed by the respective correlation coefficients: 0.13 between wind shear and ΔO_3 , *versus* 0.54 between wind shear and $\Delta \theta_e$.

A more careful analysis of the O_3 -enhancement events during the occurrence of a LLJ on April 13, 2014 (Fig. 14-a-d) provides more insights into the dynamical processes involved in the O_3 transport. The downdraft of the storm is estimated to start around 0500 h (marked by the first red line in Fig. 14), when the skewness starts an abrupt decrease to negative values. At this time, an elevated jet starts developing (Fig. 15). Nevertheless, O_3 levels only start increasing around 0520 h (second red line to the right in Fig. 14), which coincides with the increase in surface velocities associated with the LLJ. Note that a much stronger jet closer to the surface is observed in the radiosonde at 0530 h (Fig. 15).

Several similar results to those found in this study with regard to increasing turbulence intensity and wind shear during the occurrence of LLJ in fair-weather nights have been reported (Smedman et al., 1994; Karipot et al., 2008; Duarte et al., 2015). Among these stand out the results found by Hu et al. (2013) who analyzed O_3 profiles and applied a single column (1-D) chemistry model. They showed that during the passage of a nocturnal LLJ there were considerable increases of surface O_3 levels while experiencing reductions of O_3 content in the residual layer (RL). Hu et al. (2013) explained that nocturnal LLJs have the ability to connect the stable boundary layer (poor in O_3) with the RL (rich in O_3).

Figure 11: Vertical variation of the normalized wind speed ($\bar{u} U_j^{-1}$) during:(a) 22 November 2014, 0200 UTC (also shown in Fig. 2) and (b) during the observed low-level jet events in 2014.

Figure 12: Relative frequency of ozone enhancements associated with the occurrence of low-level jet events in 2014.

Figure 13: Relationship between ozone enhancements and wind shear during the occurrence of low-level jet events in 2014. The corresponding correlation coefficients are 0.13 and 0.54.

Figure 14: Time series of: a) Ozone, b) wind speed, c) skewness (Sk_w) and d) standard-deviation (σ_w) of wind vertical velocity. The vertical red lines represent the estimated times of initiation of the downdraft and the ozone enhancement.

Figure 15: Low-level jets evolution on 13 April 2014. The wind profiles from 0200-0500 h come from the sodar profiles while the profile of 0530 h corresponds to a radiosonde.

Figure 16: Surface data of (a) O_3 , (b) θ_e , (c) air density and (d) atmospheric pressure on May 07, 2014. No sudden changes are observed, and the signature of a density current is absent.

As a result, O_3 was mixed down from the RL to the surface.

However, few studies have devoted attention to the O_3 -enhancement events during storm downdrafts: in the present case, the mechanism of O_3 enhancement is different from the fair-weather cases. The additional O_3 originates above the ABL, and is transported to the surface by the storm downdrafts (as clearly shown by the vertical velocity skewness profiles). Hence, differently from the fair-weather nights when it increases mixing from the RL, here the LLJ is a manifestation of the advancing density current originating in the cold downdraft. The O_3 arrived at the surface within the core of the downdraft, which is likely at a different horizontal location from the measurement site. In this scenario, the enhanced O_3 reached the measurement site via horizontal advection within the density current. This explains the correlation between large O_3 enhancements and the presence of the LLJ.

Further confirmation between our “classical” and “non-classical” LLJ’s is the different signature of each on the surface data. We have already seen in detail, in Figures 2–3, the signature of an advancing density current. Figure 16 shows the time series of O_3 , θ_e , air density and atmospheric pressure for an LLJ detected on May 07, 2014 right before 0600 h UTC. A GOES 13 image taken at 0530 UTC confirms that no convective activity was present above the T3 site (not shown). There are no changes in any of these variables indicative of a density current. This pattern was confirmed for all cases of LLJ’s not associated with ozone enhancement events.

4. Summary and conclusions

We analyzed extensive data sets obtained in the Amazon region, near Manacapuru, Amazonas, Brazil, during 2014/2015. These data provided better and improved statistical descriptions of the relationship between convective storm downdrafts, as evidenced in physical variables related to the dynamics and thermodynamics of the atmosphere, low-level jets caused by density currents, and O_3 -enhancement events. Convective systems, which are pervasively common features in the Amazon region during the wet season, have the ability to transport O_3 from the middle troposphere to the atmospheric boundary layer. Although it has been conjectured that the type of convective activity may be related to the strength of the O_3 increase, a Ricardson number criterion applied to all enhanced O_3 events suggests that, on the average, different types of convective complexes have approximately the same ability to transport O_3 downwards.

This transport occurs through convective downdrafts, which are also related to the decrease of equivalent potential temperature near the surface as already documented in the literature. The present work provides further evidence of this mechanism by including wind profiler and radiosonde datasets. In particular, the observations of the skewness of the vertical velocity provide direct evidence of the role of the storm downdrafts in transporting tropospheric O_3 to the surface.

Other characteristic signatures of O_3 -enhancement events are high values of the horizontal wind speed, high turbulence intensity (evidenced by the σ_w sodar measurements), and large decreases of CAPE values. Regarding this last point, CAPE reduction is more strongly related to ΔO_3 than is θ_e reduction.

Moreover, during the occurrence of downdrafts, sharp reductions in the concentration of CO were also observed. The reaction between CO and O_3 is too slow in the lower atmosphere to account for the sudden drops observed, and therefore we must assign them to the same mechanism of the O_3 enhancements, namely the downdrafts bringing tropospheric air to the surface.

In the majority of the cases analyzed, low-level jets formed almost simultaneously to the O_3 increases and the reductions in equivalent potential temperature. Analysis of simultaneous surface data shows these jets to be “non-classical” (*i.e.*, their origin is different from that of fair-weather nocturnal jets), and to be associated with advancing density currents originating in the cold air downdrafts. The presence of these jets may have important consequences to the subsequent exchanges between the atmospheric surface layer and the mixed layer above, which, right after those episodes, will almost invariably (for cases with $\Delta O_3 > 8$ ppbv) occur under stable conditions (even during the day) and the increased turbulence forcing in the presence of the LLJ. However, the subsequent

turbulent exchanges of trace gases between the surface and the ABL were not analyzed in detail in the present work, and will need further scrutiny.

In addition, in environments such as the rainforest the frequent occurrences of O₃ enhancements, which reach 30 ppbv in our dataset but have been reported to be almost 40 ppbv in similar studies, can drive numerous chemical cycles. In particular, enhanced O₃ levels can contribute to the oxidation of the rainforest-emitted hydrocarbons whose reactions can generate copious amounts of hydroxyl radicals which in turn can further accelerate chemical reactions that generate precursors of secondary organic aerosols. Therefore, one key conclusion derived from this study is that, in the rainforest, one must consider the influences of both mesoscale convective storms and low-level jets originating in their downdrafts on the vertical transport of O₃ to the forest canopy to determine the oxidation of reactive gases such as rainforest-emitted hydrocarbons.

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- Andreas, E. L., Claffy, K. J., Makshtas, A. P., 2000. Low-level atmospheric jets and inversions over the western weddell sea. *Boundary-Layer Meteorol.* 97 (3), 459–486.
- Banta, R. M., Pichugina, Y. L., Brewer, W. A., 2006. Turbulent velocity-variance profiles in the stable boundary layer generated by a nocturnal low-level jet. *J. Atmos. Sci.* 63 (11), 2700–2719.
- Betts, A. K., 2002. Transport of ozone to the surface by convective downdrafts at night. *Journal of Geophysical Research* 107 (D20).
- Chase, R. J., Fuentes, J. D., Gerken, T., Wei, D., Schumacher, C., Machado, L., Santos, R. M. N., Martin, S. T., Springston, S., 2016. Convection and redistribution of tropospheric ozone in central amazonia. 15th Annual Student Conference, American Meteorological Society, 10-14 January, New Orleans, LA.
- Costantino, L., Heinrich, P., 2014. Tropical deep convection and density current signature in surface pressure: comparison between wrf model simulations and infrasound measurements. *Atmos. Chem. Phys.* 14 (6), 3113–3132.
- Duarte, H. F., Leclerc, M. Y., Zhang, G., Durden, D., Kurzeja, R., Parker, M., Werth, D., 2015. Impact of nocturnal low-level jets on near-surface turbulence kinetic energy. *Boundary-Layer Meteorology* 156 (3), 349–370.
- Finlayson-Pitts, B. J., Pitts, J. N., 2000. *Chemistry of the Upper and Lower Atmosphere: Theory, Experiments, and Applications*. Academic Press, London.
- French, A. J., Parker, M. D., 2012. Observations of mergers between squall lines and isolated supercell thunderstorms. *Weather and Forecasting* 27 (2), 255–278.

481 Fuentes, J. D., Lerdau, M., Atkinson, R., Baldocchi, D., Bottenheim, J. W., Ciccioli, P., Lamb,
482 B., Geron, C., Gu, L., Guenther, A., et al., 2000. Biogenic hydrocarbons in the atmospheric
483 boundary layer: a review. *Bull. Amer. Meteorol. Soc.* 81 (7).

484 Garstang, M., Scala, J., Greco, S., Harriss, R., Beck, S., Browell, E., Sachse, G., Gregory,
485 G., Hill, G., Simpson, J., Tao, W.-K., Torres, A., 1988. Trace gas exchanges and convective
486 transports over the amazonian rain forest. *Journal of Geophysical Research: Atmospheres*
487 93 (D2), 1528–1550.

488 Gerken, T., Wei, D., Chase, R. J., Fuentes, J. D., Schumacher, C., Machado, L. A., Andreoli,
489 R. V., Chamecki, M., de Souza, R. A. F., Freire, L. S., Jardine, A. B., Manzi, A. O., dos Santos,
490 R. M. N., von Randow, C., dos Santos Costa, P., Stoy, P. C., Tóta, J., Trowbridge, A. M.,
491 2016. Downward transport of ozone rich air and implications for atmospheric chemistry in
492 the amazon rainforest. *Atmospheric Environment* 124, Part A, 64 – 76.

493 Grant, D. D., Fuentes, J. D., DeLonge, M. S., Chan, S., Joseph, E., Kucera, P., Ndiaye, S. A.,
494 Gaye, A. T., 2008. Ozone transport by mesoscale convective storms in western senegal. *At-*
495 *mospheric Environment* 42 (30), 7104 – 7114.

496 Guenther, C. C., 2006. Estimates of global terrestrial isoprene emissions using megan (model of
497 emissions of gases and aerosols from nature). *Atmos. Chem. Phys.* 6.

498 Halverson, J. B., Rickenbach, T., Roy, B., Pierce, H., Williams, E., 2002. Environmental charac-
499 teristics of convective systems during trmm-lba. *Mon. Wea. Rev.* 130 (6), 1493–1509.

500 Hu, X.-M., Fuentes, J. D., Zhang, F., 2010a. Downward transport and modification of tropo-
501 spheric ozone through moist convection. *Journal of atmospheric chemistry* 65 (1), 13–35.

502 Hu, X.-M., Fuentes, J. D., Zhang, F., 2010b. Downward transport and modification of tropo-
503 spheric ozone through moist convection. *Journal of Atmospheric Chemistry* 65 (1), 13–35.

504 Hu, X.-M., Klein, P. M., Xue, M., Zhang, F., Doughty, D. C., Forkel, R., Joseph, E., Fuentes,
505 J. D., 2013. Impact of the vertical mixing induced by low-level jets on boundary layer ozone
506 concentration. *Atmospheric Environment* 70, 123–130.

507 Iribarne, J. V., Godson, W. L., 1986. *Atmospheric Thermodynamics*. D. Reidel, Dordrecht.

508 Jardine, A. B., Jardine, K. J., Fuentes, J. D., Martin, S. T., Martins, G., Durgante, F., Carneiro, V.,
509 Higuchi, N., Manzi, A. O., Chambers, J. Q., 2015. Highly reactive light-dependent monoter-
510 penes in the amazon. *Geophys. Res. Lett.* 42 (5), 1576–1583.

511 Karipot, A., Leclerc, M. Y., Zhang, G., Lewin, K. F., Nagy, J., Hendrey, G. R., Starr, G., 2008.
512 Influence of nocturnal low-level jet on turbulence structure and co2 flux measurements over a
513 forest canopy. *Journal of Geophysical Research: Atmospheres* 113 (D10), D10102.

514 Kirchhoff, V. W. J. H., da Silva, I. M. O., Browell, E. V., 1990. Ozone measurements in amazonia:
515 Dry season versus wet season. *Journal of Geophysical Research: Atmospheres* 95 (D10),
516 16913–16926.

517 Martin, S. T., Artaxo, P., Machado, L. A. T., Manzi, A. O., Souza, R. A. F., Schumacher, C.,
518 Wang, J., Andreae, M. O., Barbosa, H. M. J., Fan, J., Fisch, G., Goldstein, A. H., Guenther,
519 A., Jimenez, J. L., Poschl, U., Silva Dias, M. A., Smith, J. N., Wendisch, M., 2015. Introduc-
520 tion: Observations and modeling of the green ocean amazon (goamazon2014/5). *Atmospheric*
521 *Chemistry and Physics Discussions* 15, 30175–30210.

522 Mitchell, J. F., 1989. The “greenhouse” effect and climate change. *Rev. Geophys.* 27 (1), 115–
523 139.

524 Rafkin, S. C. R., Barth, E. L., 2015. Environmental control of deep convective clouds on titan:
525 The combined effect of cape and wind shear on storm dynamics, morphology, and lifetime.
526 *Journal of Geophysical Research: Planets* 120 (4), 739–759.

- 527 Rotunno, R., Klemp, J. B., Weisman, M. L., 1988. A theory for strong, long-lived squall lines.
528 *Journal of the Atmospheric Sciences* 45 (3), 463–485.
529 URL [http://dx.doi.org/10.1175/1520-0469\(1988\)045<0463:ATFSL>2.0.CO;2](http://dx.doi.org/10.1175/1520-0469(1988)045<0463:ATFSL>2.0.CO;2)
- 530 Sahu, L. K., Lal, S., 2006. Changes in surface ozone levels due to convective downdrafts over
531 the bay of bengal. *Geophysical Research Letters* 33 (10), L10807.
- 532 Santana, R. A. S. d., Tota, J., Santos, R. M. N. d., Vale, R. S. d., 12 2015. Estabilidade e estrutura
533 da turbulência sob a influência de jatos de baixos níveis noturnos no sudoeste da amazônia.
534 *Revista Brasileira de Meteorologia* 30, 405–414.
- 535 Scala, J. R., Garstang, M., Tao, W.-k., Pickering, K. E., Thompson, A. M., Simpson, J., Kirch-
536 hof, V. W. J. H., Browell, E. V., Sachse, G. W., Torres, A. L., Gregory, G. L., Rasmussen,
537 R. A., Khalil, M. A. K., 1990. Cloud draft structure and trace gas transport. *Journal of Geo-*
538 *physical Research: Atmospheres* 95 (D10), 17015–17030.
- 539 Sigler, J. M., Fuentes, J. D., Heitz, R. C., Garstang, M., Fisch, G., 2002. Ozone dynamics and de-
540 position processes at a deforested site in the amazon basin. *AMBIO: A Journal of the Human*
541 *Environment* 31 (1), 21–27.
- 542 Smedman, A.-S., Tjernström, M., Högström, U., 1994. The near-neutral marine atmospheric
543 boundary layer with no surface shearing stress: A case study. *American Meteorological Soci-*
544 *ety* 51 (23), 3399–3411.
- 545 Weisman, M. L., Klemp, J. B., 1982. The dependence of numerically simulated convective
546 storms on vertical wind shear and buoyancy. *Monthly Weather Review* 110 (6), 504–520.
- 547 Weisman, M. L., Rotunno, R., 2004. “A theory for strong long-lived squall lines” revisited.
548 *Journal of the Atmospheric Sciences* 61 (4), 361–382.
549 URL [http://dx.doi.org/10.1175/1520-0469\(2004\)061<0361:ATFSL>2.0.CO;2](http://dx.doi.org/10.1175/1520-0469(2004)061<0361:ATFSL>2.0.CO;2)
- 550 Wissmeier, U., Goler, R., 2009. A comparison of tropical and midlatitude thunderstorm evolution
551 in response to wind shear. *Journal of the Atmospheric Sciences* 66 (8), 2385–2401.
- 552 Wofsy, S. C., McElroy, M. B., 1974. Hox, nox, and clox: Their role in atmospheric photochem-
553 istry. *Canadian Journal of Chemistry* 52 (8), 1582–1591.