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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.

1. Introduction

Ozone (O₃) 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), O₃ influences numerous chemical cycles (Wofsy and McElroy, 1974). For example, O₃ reacts with soil-emitted nitric oxide (NO) to form nitrogen dioxide (NO₂) and molecular oxygen (O₂), i.e., O₃ + NO \rightarrow NO₂ + O₂. The photolysis of NO₂ generates NO and atomic oxygen (O) which readily combines with O₂ to reform O₃ in the presence of a third molecule such as nitrogen (N₂). Also, O₃ can undergo photolysis to generate O₂ and electronically excited oxygen atoms (O(¹D)), that is, O₃ + $h_{\nu} \rightarrow$ O(¹D) + O₂. In rich water vapor (H₂O) environments such as over the Amazon rainforest, the O(¹D) rapidly combines with H₂O to produce hydroxyl radicals (OH) (Finlayson-Pitts and Pitts,

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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 (CH₄) (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

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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, ground-level O₃ can suddenly increase in response to the transport of air parcels from the middle of the troposphere where O₃ levels are considerably higher than at the surface (Kirchhoff et al., 1990). Ground-level O₃ enhancements caused by storms depend on several factors, including intensity of atmospheric convection, storm propagation speed, origin of downward moving air parcels, and time of the day (Betts, 2002; Sahu and Lal, 2006; Grant et al., 2008; Hu et al., 2010b; Gerken et al., 2016). The O₃ enhancements can be as much as 30 ppbv.

With the exception of the study by Gerken et al. (2016) that included almost one year of field observations over the rainforest of Brazil, most studies report storm-related O₃ enhancements for brief field campaigns.

The processes governing O₃ enhancements in the atmospheric boundary layer over the rainforest, associated with storms, are crucial to understand the chemical cycles involved with the formation of oxidants and aerosols. For example, the Amazon rainforest emits large amounts of isoprene, monoterpenes, and sesquiterpenes (Guenther, 2006; Jardine et al., 2015). Such gases react with O₃ to form free radicals (e.g., OH) and precursors of secondary organic aerosols (Fuentes et al., 2000). Therefore, one objective of the present study is to investigate the atmospheric thermodynamic (e.g., convective available potential energy (CAPE)) and dynamic (e.g., standard deviation (σ_w) and skewness (Sk_w) of the vertical velocity) conditions associated with storm-generated O₃ enhancements in the Amazon rainforest. A second objective is to determine the magnitude and the frequency of ground-level O₃ enhancements related to the appearance of "non-classical" lowlevel jets (LLJ's) — in the sense that they are different from classical nocturnal LLJ's (see for example Banta et al., 2006) — that develop in response to those storms. For this purpose, air chemistry and meteorological data sets, obtained as part of an unprecedented year-long measurement campaign in the Amazon region within the context of the GoAmazon project (Martin et al., 2015), are analyzed and interpreted, and the LLJ's associated with O₃ enhancements are identified with advancing density currents originating from the storm cold downdrafts.

2. Methods

2.1. Experimental site

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

$$CAPE = \int_{z_{\rm f}}^{z_{\rm n}} g \frac{T_{\nu,sa} - T_{\nu}}{T_{\nu}} dz$$
 (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); $T_{\nu,sa}$ is the virtual temperature profile of a saturation adiabat starting at the lifting condensation level (LCL), and T_{ν} 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-min 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 Δ CAPE 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 O₃-enhancement events. For this, National Oceanic and Atmospheric Administration (NOAA) Geostationary Operational Environmental Satellite System 13 (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°08.98′ S, 59°59.48′ 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)

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