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Key Point:

 Impacts of a tropical cyclone are seen in tropospheric ozone at midlatitudes

Supporting Information:

• Texts S1 and S2, Figures S1 and S2, and Figure S3

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Signature of a tropical Pacific cyclone in the composition of the upper troposphere over Socorro, NM

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Abstract We present a case study based on balloon-borne ozone measurements during the SouthEast American Consortium for Intensive Ozonesonde Network Study in August–September 2013. Data from Socorro, NM (34°N, 107°W) show a layer of anomalously low ozone in the upper troposphere (UT) during 8–14 August. Back trajectories, UT jet analyses, and data from the Microwave Limb Sounder (MLS) on the Aura satellite indicate that this feature originated from the marine boundary layer in the eastern/central tropical Pacific, where several disturbances and one hurricane (Henriette) formed within an active region of the Intertropical Convergence Zone in early August 2013. The hurricane and nearby convection pumped boundary layer air with low ozone (20–30 ppbv) into the UT. This outflow was advected to North America 3–5 days later by a strong subtropical jet, forming a tongue of low ozone observed in MLS fields and a corresponding layer of low ozone in Socorro vertical profiles.

1. Introduction

Ozone in the upper troposphere (UT, from about 8 km altitude to the tropopause) is one of the more important trace gases for the chemistry and radiative balance in this critical region of the atmosphere. The overall radiative forcing by tropospheric ozone displays large spatial and temporal variability [*Mickley et al.*, 1999, 2004], which is driven in large part by the variability in UT ozone. The photochemical lifetime of ozone in the UT is influenced by many factors, including the availability of sunlight, nitrogen oxides, and other reactive gases, but under most conditions the timescale for production or destruction of ozone above the surface boundary layer ranges from ~10 days to a few weeks [*Liu et al.*, 1980; *Logan et al.*, 1981]. The quasi-horizontal transport of trace gases can occur on much shorter timescales, especially when it occurs within persistent wind patterns such as UT jets [e.g., *Manney et al.*, 2014]. This contrast in timescales allows for the long-range transport of UT ozone far from source regions [e.g., *Sudo and Akimoto*, 2007]. Studies by *Berntsen et al.* [1999] and *Jacob et al.* [1999] examined the importance of Asian emissions on North American air quality via transport of ozone and other pollutants in the free troposphere over the Pacific Ocean, and similar efforts focused on transport across the Atlantic Ocean from North America to Europe [*Stohl and Trickl*, 1999; *Li et al.*, 2002].

UT ozone can also be significantly enhanced (>100 ppbv) on synoptic scales by irreversible transport of stratospheric air into the upper troposphere during episodes of stratosphere-troposphere exchange (STE) [*Gettelman et al.*, 2011, and references therein]. Gauging the global impact of STE on UT ozone directly from observations has been limited by the scarcity of measurements with high vertical resolution and dense spatial/temporal sampling [*Tang and Prather*, 2012]. *Thompson et al.* [2007a] estimated the contribution from STE to the summertime UT ozone budget over northeastern North America at ~25% based on data from the Intercontinental Chemical Transport Experiment Ozonesonde Network Study. *Pan et al.* [2014] examined aircraft data that captured a significant transport event of stratospheric ozone down to the UT associated with a mesoscale convective system over the midcontinental United States.

While the long-range horizontal transport of tropospheric ozone generally occurs over timescales of days to a week, UT ozone can be affected even more rapidly by the vertical transport in deep convection. *Kley et al.* [1996] observed very low O_3 in the upper troposphere over the equatorial Pacific that was attributed to rapid lofting of

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Figure 1. All (left) ozone and (right) temperature vertical profiles from Socorro, 8 August to 23 September 2013. Those ozone profiles taken between 8 and 14 August 2013 are plotted in red. All others are plotted in black.

boundary layer air within local deep convection (cloud tops in the UT) or to lateral outflow from nearby regions of active convection. More generally, deep convection has been shown to have a substantial impact on mean UT ozone and its seasonal variability over a wide range of spatial scales [e.g., *Thompson et al.*, 2007b, 2010; *Folkins et al.*, 2002; *Petropavlovskikh et al.*, 2010], with the largest impacts generally occurring in the tropics [*Livesey et al.*, 2013], or at higher latitudes within active monsoon regions [*Randel and Park*, 2006].

Here we present an analysis of UT ozone measurements over a continental site in North America that carry the signature of deep maritime convection over the eastern and central tropical Pacific. The convection was part of one hurricane and several tropical disturbances that occurred between 3000 and 5000 km away from and 3 to 5 days prior to the continental ozone measurements.

2. Data

Ozone vertical profile measurements were made over Socorro, New Mexico (34°N, 107°W, 1.42 km altitude), in August 2013 during SEACIONS (SouthEast American Consortium for Intensive Ozonesonde Network Study). SEACIONS soundings were made nearly daily from 8 August to 20 September 2013, from seven North American stations (Boulder, CO; Houston, TX; Huntsville, AL; Idabel, OK; Socorro, NM; St. Louis, MO; and Tallahassee, FL). In total, 209 soundings were made; refer to http://croc.gsfc.nasa.gov/seacions. One of the objectives of SEACIONS was to support the SEAC⁴RS (Studies of Emissions and Atmospheric Composition, Clouds, and Climate Coupling by Regional Surveys) mission, which focused on deep convection and its role in the redistribution of pollutant emissions and aerosols throughout the troposphere.

Ozonesonde data were obtained using an in situ sensor [Komhyr, 1986; Komhyr et al., 1995] flown on a balloon package that included a standard meteorological radiosonde for simultaneous measurements of pressure, temperature, relative humidity, and winds. The ozone instrument consists of a Teflon air pump and an electrochemical ozone sensor (electrochemical concentration cell (ECC)) with two platinum electrodes in separate cells of potassium iodide solutions with different concentrations. Ambient air is pumped through one cell, and the presence of ozone drives chemical reactions that give rise to a small (microampere) current between the electrodes. Output from the ECC is interfaced to the meteorological radiosonde, which transmits all of the ozone and meteorological data to a ground receiving station during the ~2 h balloon ascent up to about 30 km maximum altitude. The precision in ozone mixing ratios in the troposphere is 3-5% (1 σ), the absolute accuracy is about 10%, and the combined effect from the sensor time response in the upper troposphere (~25 s) and the balloon ascent rate (4–5 m s⁻¹) produces an effective vertical resolution of 100–125 m [Hassler et al., 2014, and references therein].

3. Ozone Vertical Profiles: Signatures of Tropical Pacific Convection

A total of 30 ozone vertical profiles were obtained from Socorro during the period 8 August to 23 September 2013 (Figure 1, left). Ozone mixing ratios near the surface ranged from about 40 to 60 ppbv, but at higher altitudes in the UT, ozone mixing ratios were much larger and displayed higher variability, with values between 50 and 100 ppbv. A major component of this variability was due to the presence of laminar structures in the UT. These laminae had typical vertical thicknesses between 0.2 and 2 km, with peak amplitudes in ozone between 0% and + -30% relative to the mean. Some of these structures can be discerned from the individual vertical profiles in Figure 1. In contrast, temperatures profiles at Socorro (Figure 1, right) exhibited smaller variability in the UT, and a tropopause altitude that was consistently near 16 km.



Figure 2. (left) Socorro ozonesonde vertical profile measured on 12 August (solid black curve). Also shown is the mean Socorro ozone profile during the SEACIONS period 8 August to 23 September (solid blue) along with the range of + - 1 standard deviation at each altitude (dashed blue), and an MLS/OMI GEOS assimilated profile interpolated to the location of Socorro for 12 August (diamonds). (right) Colors indicate deviations of ozone vertical profiles from the mean, in units of standard deviation, for the period 8 August to 16 August. No data were obtained on 11 August.

Laminae in trace gas profiles have been observed at all latitudes in the UT and lower stratosphere for many years [e.g., *Ehhalt et al.*, 1983; *Reid and Vaughan*, 1991]. Most of these structures in ozone vertical profiles can be traced to dynamical processes such as gravity or Rossby waves [e.g., *Teitelbaum et al.*, 1994; *Grant et al.*, 1998; *Manney et al.*, 1998; *Thompson et al.*, 2011], convection [e.g., *Avery et al.*, 2010; *Morris et al.*, 2010; *Petropavlovskikh et al.*, 2010], or intrusions of air masses with high ozone from the stratosphere [e.g., *Thompson et al.*, 2007a; *Olsen et al.*, 2013]. Many transient laminae have limited horizontal scales, leading to spatial ozone variability and the appearance of tongues or filaments in ozone fields [e.g., *Bowman et al.*, 2007].

During 8–14 August, a series of low-ozone laminae were observed in the UT over Socorro. Minimum mixing ratios in these laminae were up to 2 standard deviations below the mean value at that altitude. Figure 2 (left) shows the Socorro mean ozone profile and its standard deviation. Also displayed in Figure 2 is the profile from 12 August, which shows a large amplitude (>1 σ) layer of low ozone between 9 and 12 km. Figure 2 (right) shows results from eight profiles obtained during 8–16 August, expressed in units of standard deviations from the mean. A low ozone lamina was present near ~13 km on 8 August, and similar laminae were observed at successively lower altitudes down to about 9 km on 14 August. After this time, low-ozone structures were not observed except on isolated days, and the thicknesses and amplitudes were generally not as large as those observed during 8–14 August.

Evidence from satellite measurements and back trajectory calculations show that these structures originated from the marine boundary layer over the eastern/central tropical Pacific Ocean. Figure 3 outlines the dynamical processes along with the evolution of UT ozone during part of the above time period. Figure 3 (top left) shows ice water content (IWC) at the 215 hPa pressure level measured by the Microwave Limb Sounder (MLS) instrument on the Aura satellite [*Livesey et al.*, 2015, and references therein] on 8 August 2013. Several tropical disturbances and one hurricane formed along an active region of the ITCZ (Intertropical Convergence Zone) in the eastern/central Pacific during the first 2 weeks of August 2013, as evidenced by the large IWC values from high cloud tops in Figure 3. Hurricane Henriette was a small/medium cyclone that formed over the eastern Pacific and reached category 2 intensity (on the Saffir-Simpson Hurricane Wind Scale) on 6 August before weakening and moving into the central Pacific basin on 9 August [*Berg and Powell*, 2014]. The storm ultimately dissipated several hundred miles southwest of Hawaii. This hurricane, along with a series of other tropical disturbances both prior to, and in the wake of the cyclone, pumped marine boundary layer air with low ozone (20–30 ppbv) into the UT over a large region of active convection.

Figure 3 (bottom left) shows UT ozone measured by MLS [*Livesey et al.*, 2015] interpolated to the 345 K potential temperature surface (roughly 10–11 km). The MLS data show a large region of low UT ozone over the

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Figure 3. Processes associated with the observation of low UT ozone over Socorro during the period 8–16 August. (top left) lce water content at 215 hPa on 8 August, showing high clouds from hurricane Henriette near 17°N, 222°W and from nearby tropical disturbances in the eastern/central tropical Pacific. Black contours show wind speeds from a jet/tropopause analysis [*Manney et al.*, 2011], with wind contours every 10 m/s starting at 30 m/s. (bottom left) MLS ozone mixing ratios at 345 K (~10.5 km) on 8 August, showing low ozone from convective lofting of boundary layer air into the UT. Black symbols indicate the track of individual MLS measurements for that day. (bottom right) Cluster of 5 day back trajectories run from 12 August (blue), initialized at 345 K and at locations ±0.5° around the Socorro site. Also shown is the path of Henriette (red symbols; 8 August in blue). (top right) MLS ozone field at 345 K on 12 August, showing low UT ozone advected from the Pacific to SW North America. The location of Socorro is indicated by a black diamond. Wind speed contours are shown in black.

tropical Pacific, along and north of the area of active convection. The very low ozone mixing ratios at 345 K are consistent with surface values observed over a broad area of the Pacific at that time. Measurements of ozone from Mauna Loa Observatory, Hawaii [*Oltmans and Levy*, 1994; *Lin et al.*, 2014], indicate ozone levels between 20 and 40 ppbv in primarily boundary layer air (sampled in the late afternoon) during the first 2 weeks of August 2013, and surface measurements from Samoa are in the range 15–25 ppbv. Ozonesonde measurements from Hilo, Hawaii, on 2 and 7 August clearly depict low ozone (20–30 ppbv) in the upper troposphere between 10 and 14 km, indicative of convective lofting of low ozone from the marine boundary layer. In contrast, surface ozone measured at Socorro in early August was consistently near 50 ppbv, and local convection therefore could not have produced the low-ozone laminae seen in Figure 2.

Back trajectories initialized over Socorro on 12 August (Figure 3, bottom right) show that air parcels at 345 K originated from the region of low UT ozone in the eastern/central tropical Pacific. Most of this transport occurred along a strong subtropical UT jet that is identified by the black wind contours on the IWC map in Figure 3. The jet transported a tongue of low ozone northeastward from the tropical Pacific, as seen in the MLS UT ozone field on 12 August (Figure 3, top right). The exact shape and extent of this low-ozone tongue may not be mapped precisely using the satellite data, given the spatial sampling frequency, horizontal averaging (~350 km), and vertical resolution (~3.5 km) of the MLS UT ozone measurements [Livesey et al., 2015]. Nevertheless, the MLS UT ozone data are in very good agreement with the Socorro profile from 12 August (see supporting information) and thus strongly support the conclusion that the low UT ozone was transported over southwestern North America from the eastern/central tropical Pacific. Further support is provided by comparisons with assimilated ozone profiles from MLS and total column ozone from the Ozone Monitoring Instrument (OMI). This assimilated ozone product is produced by the Global Modeling and Assimilation Office (GMAO) by ingesting OMI v8.5 total column ozone and MLS v3.3 ozone profiles into a modified version of the Goddard Earth Observing System version 5.7.2 (GEOS 5.7.2) assimilation system [Wargan et al., 2015]. Figure 2 includes an ozone vertical profile interpolated to the location of Socorro for 12 August 2013, which shows a layer of low UT ozone similar to that measured by the ozonesonde.

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Figure 4. (top) Vertical profiles of ozone and relative humidity measured over Socorro on 9 August 2013 (solid) and over Hilo on 7 August 2013 (dashed). (bottom) MLS measurements of relative humidity with respect to ice at 215 hPa on 9 August, along with wind contours at this level.

The back trajectory paths were calculated using a trajectory code [Lawrence et al., 2015] driven by winds and diabatic heating rates from the Modern-Era Retrospective analysis for Research and Applications (MERRA) reanalysis [Rienecker et al., 2011]. For the results shown in Figure 3, a cluster of 25 trajectories were initialized on the 345 K potential temperature surface in a 1°×1° latitude/longitude square region centered over Socorro on 12 August. The divergence of trajectory paths becomes apparent after going back about 1 day, and going back to 5 days, the spread in locations over the eastern/central tropical Pacific provides a rough measure of the uncertainty in the origin of air masses sampled over Socorro during the period 11-13 August. Thus, while it is not possible to pinpoint the exact origins of the observed low-ozone laminae back to hurricane Henriette, the evidence strongly suggests that convection from this storm played a major role in shaping the composition of the UT over Socorro during this period.

Previous studies of UT composition from aircraft have shown low ozone in convective detrainment regions of tropical cyclones [*Cairo et al.*, 2008, and references therein] and near tropical convective disturbances [e.g., *Pickering et al.*, 2001]. In addition, climatologies of ozonesonde profiles from tropical maritime sites contain convective signatures in UT ozone [*Thompson et al.*, 2011; *Paulik and Birner*, 2012]. Well-defined bubbles of anomalously low UT equatorial ozone were analyzed by *Petropavlovskikh et al.* [2010], and they concluded that these bubbles originated from distant areas of deep tropical convection that occurred at least 5 days prior to the ozone observations.

In contrast to the above low-latitude observations, there are few studies showing that UT ozone over a midlatitude continental site can be significantly impacted by tropical maritime convection. *Vogel et al.* [2014] suggested, based on measurements of O_3 and other trace gases in the lower stratosphere over Northern Europe in September 2012, that boundary layer air from Southeast Asia was lofted rapidly within a typhoon and subsequently transported to the high-latitude lowermost stratosphere over a period of many weeks (up to 40 days). They showed that eastward eddy shedding from the Asian monsoon anticyclone was an important process for this transport pathway into the lowermost stratosphere. For the case study presented here, the vertical transport mechanism is similar, but the quasi-horizontal transport pathway to the UT over southwestern North America is more rapid and direct.

One interesting aspect of these series of measurements was the presence of low relative humidity associated with low ozone. In light of abundant evidence that the overall effect of tropical deep convection is to moisten the UT [e.g., *Su et al.*, 2006], we might have expected higher humidity to accompany the Socorro low-ozone observations. Figure 4 shows a map of relative humidity with respect to ice (RHi) at 345 K measured by MLS [*Livesey et al.*, 2015] on 9 August, along with vertical profiles of ozone and radiosonde relative humidity from Socorro on 9 August and from Hilo on 7 August. The MLS RHi map shows a large area of dry air over the Pacific Ocean to the north of hurricane Henriette, extending northeastward to western North America. The relative humidity at Hilo and Socorro indicates a layer of dry air that coincides with the region of low ozone in the upper troposphere. Dry air layers were also observed on other days during 8–14 August when low ozone was observed at Socorro.

A detailed analysis of dynamical processes affecting cyclone outflow patterns and the thermodynamics of deep convective moistening and drying tendencies is beyond the scope of this study. However, it is important to note that the UT wind field associated with tropical cyclones is anticyclonic and typically contains an outflow layer between 300 and 100 hPa [Black and Anthes, 1971]. The outflow often takes the form of a jet linking the cyclone with midlatitude westerlies [Merrill, 1988]. Ray and Rosenlof [2007] analyzed satellite water vapor fields in the vicinity of tropical cyclones and found that cyclones hydrate the UT in the vicinity of the storms by up to 50% above the monthly mean humidity. However, the moist region was found immediately east of the cyclone eye and to a lesser extent to the south and southwest of the eye. On the northwest and northern quadrants, corresponding to the origins of the many of the trajectories in Figure 3 with respect to Henriette, the air was relatively drier but likely not enough to explain the very low (5-10%) relative humidity observed over Socorro. On the other hand, relative humidity is not a conserved tracer and can be significantly modified by small changes in temperature. Our back trajectory analysis indicates sinking motion and a 5–10 K warming for air parcels transported in the subtropical jet region, consistent with the details of the MLS RHi fields and the Hilo and Socorro relative humidity values shown in Figure 4. Relative humidity simulations (see supporting information) show that these dynamically induced effects are sufficient to explain the low relative humidity observed at Socorro.

4. Conclusions

Ozone vertical profiles over Socorro, NM taken during 8–14 August as part of the SEACIONS campaign showed layers of anomalously low ozone in the UT between 8 and 14 km. These ~1–3 km thick laminae contained minimum ozone mixing ratios that were $1-2\sigma$ lower than the mission mean, were less than the range of values seen locally at the surface, and were most consistent with values found in the remote marine boundary layer. Trajectory calculations show that these laminae can be traced back 3–5 days to the eastern/central tropical Pacific, a distance of up to 5000 km. Satellite data and analysis of meteorological fields demonstrate that the observed laminae were part of a tongue of low-ozone air that was drawn northeastward by a strong subtropical Pacific jet stream. The low UT ozone was a result of deep convection within hurricane Henriette and other nearby tropical disturbances. The relative humidity within these UT ozone laminae was low, however, which can be attributed to descending motion and significant warming (5–10 K) during transport from the central Pacific.

The temporally limited ozone data set from Socorro does not show any other distinct signatures of tropical convection, so it is unclear how frequently such episodes may occur. In terms of spatial dimensions, the MLS ozone fields indicate that the tongue of low ozone seen on 12 August did not extend significantly north or east of Socorro, and persistent low-ozone laminae were not consistently observed at any of the other SEACIONS stations during 8–14 August. Based on a single case study such as this, it is not possible to quantify the overall importance of lofting and long-range transport of tropical marine boundary layer air in controlling the mean amount of UT ozone over North America. Further work is needed to identify reliable diagnostics for these kinds of events and to quantify their frequency and intensities from longer term data records.

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Acknowledgments

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