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Analysis of satellite remote sensing observations of low ozone events in the tropical upper troposphere and links with convection

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[1] Satellite observations from three instruments (Microwave Limb Sounder, Optical Spectrograph and Infrared Imaging System, and Atmospheric Chemistry Experiment Fourier Transform Spectrometer) reveal coherent patterns of low ozone events (<20 ppb) in the tropical upper troposphere. Using a chemical transport model (GEOS-Chem), we find that these events result from deep convective processes that rapidly transport air with low ozone concentrations from the marine boundary layer. These events occur with greater frequency over the tropical South Pacific warm pool, which is consistent with ozonesonde observations. The satellite observations indicate spatial shifts in the frequency of low ozone events that we attribute to changes in convection. As the location of the warm pool shifts eastward during El Niño events, the location of the most frequent low ozone events in the satellite record follows. Mapping of low ozone events over time reveals eastward propagating systems resembling the Madden-Julian Oscillation. These observations and analyses strengthen the link between deep convection and ozone concentrations in the tropical upper troposphere.
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1. Introduction

[2] Ozone (O_3) is an important contributor to upper tropospheric oxidation processes and radiative forcing [*Lacis et al.*, 1990; *Thompson*, 1992]. Upper tropospheric ozone is greatly affected by convection, in addition to the chemical production and loss mechanisms [*Folkins et al.*, 2002; *Sekiya and Sudo*, 2012]. Dynamic processes in the tropics vary on weekly, seasonal, and interannual time scales leading to variability in ozone concentrations. Strengthening scientific understanding about the link between convection and upper tropospheric ozone concentrations is essential to understand ozone and its variability.

[3] The tropical marine boundary layer is a region with intense solar radiation and high humidity. Under these conditions, an O_3 molecule is easily photolyzed, and the resulting excited oxygen atom readily reacts with water vapor, leading to a net loss of O_3 [*Johnson et al.*, 1990]. This leads to O_3 concentrations that reach values as low as a few ppb in the clean tropical marine boundary layer [*Kley et al.*, 1996]. Deep convection in the tropics is believed to bring this air with low ozone concentrations to the upper troposphere causing reduced ozone columns [*Folkins et al.*, 2002]. The resulting depletion in ozone concentrations has been observed by ozonesondes in a few convectively active regions [*Kley et al.*, 1996; *Solomon et al.*, 2005]. However, the spatial extent of these events remains weakly quantified.

[4] Interannual variability of convection is largely driven by the El Niño–Southern Oscillation (ENSO) [*Sekiya and Sudo*, 2012]. During El Niño events, warming of the central and eastern Pacific Ocean leads to significant changes in the strength and distribution of convective events as a result of the strong link between convection and surface temperatures. A recent study by *Oman et al.* [2013] has used satellite observations and a chemistry-climate mode to describe the sensitivity of mean upper tropospheric ozone concentrations to ENSO.

[5] The Madden-Julian Oscillation (MJO) is the dominant source of seasonal variability in the tropics [*Madden and Julian*, 1994]. The MJO is identified by extensive regions of enhanced convection that form over the Indian Ocean and propagate eastward along the equator with a period of 40–50 days. Ozone anomalies connected to the MJO have been previously observed in O_3 column data from satellites [*Ziemke and Chandra*, 2003; *Tian et al.*, 2007] but, to our knowledge, have not been observed in the upper troposphere.

[6] Scientific understanding of the effects of convection on ozone in the upper troposphere has been inhibited by the paucity of observations. Ozonesondes have provided a glimpse into these connections at a few locations [*Kley et al.*, 1996; *Solomon et al.*, 2005; *Lee et al.*, 2010]. Satellite observations of column ozone yield hints about the global nature of these processes [*Shiotani*, 1992; *Ziemke et al.*, 2010; *Chae et al.*, 2011], but without specificity to the upper troposphere. Here we apply satellite observations of upper tropospheric ozone to detect convectively driven low ozone events from space at unprecedented spatial and temporal scales and to analyze these events with a chemical transport model.

Additional supporting information may be found in the online version of this article.

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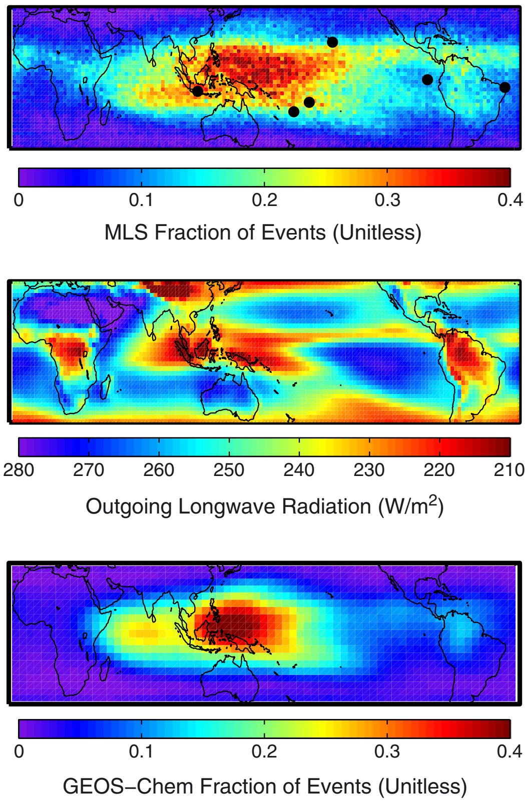

Figure 1. (Top) Frequency of low ozone events (<20 ppb) at 215 hPa observed by the MLS satellite instrument over the years 2004–2012. Black dots indicate ozonesonde locations. (Middle) Observations of outgoing longwave radiation (OLR), for which low values indicate regions of deep convection. (Bottom) Frequency of low ozone events in a GEOS-Chem simulation for 2004–2010 with random Gaussian noise added to model output to simulate MLS measurement precision.

2. Data

[7] We use satellite observations in three spectral regions (microwave, ultraviolet-visible, and midinfrared) to observe low ozone events and reduce the chance of artifacts affecting our conclusions. The Aura Microwave Limb Sounder (MLS) is a thermal emission microwave limb sounding instrument launched in July 2004. MLS makes 3500 profile measurements from the upper troposphere to the mesosphere with a horizontal spacing of 165 km daily. In this study, we use version 3.3 of the MLS data processing algorithms for retrievals between August 2004 and January 2012 [*Livesey et al.*, 2011]. The wavelengths of the measured radiation are such that thin and moderate clouds do not affect the retrievals; however, thick clouds may affect the retrievals. We ignore any measurements which may have been influenced by clouds through the recommended screening processes using the MLS data quality flags. Comparisons between MLS O_3 and ozonesondes show the best agreement (better than 10%) in the tropical upper troposphere at 215 hPa with small positive biases (<10 ppb over the Pacific) but indicate larger biases at other pressure levels [*Livesey et al.*, 2012].

[8] The Optical Spectrograph and Infrared Imaging System (OSIRIS) was launched in 2001 and measures scattered sunlight in the limb with wavelengths ranging from ultraviolet to infrared (280–800 nm) [*Llewellyn et al.*, 2004; *McLinden et al.*, 2012]. Daily observations are made between 6 and 60 km with a horizontal resolution of 500–1000 km and a vertical resolution of 2 km. This study uses the SaskMART ozone product version 5-07 for orbits occurring between October 2001 and June 2012 [*Degenstein et al.*, 2009]. Annual mean OSIRIS O_3 concentrations agree with in situ observations to within 5% in the tropical upper troposphere, although seasonal biases are larger and high-altitude clouds may cause retrieval errors [*von Savigny et al.*, 2005; *Cooper et al.*, 2011].

[9] The Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) is a solar occultation instrument launched in August 2003 measuring infrared radiation (750–4400 cm^{-1}), providing a few hundred ozone profiles per year in the tropics [*Bernath*, 2006]. Ozone observations are made between the cloud tops and 95 km with a vertical resolution of ~3 km, and profiles are retrieved using a nonlinear least squares fitting method [*Boone et al.*, 2005]. ACE-FTS version 3.0 profiles for measurements between February 2004 and September 2010 are used for this work. Annual mean ozone profiles generally agree with in situ observations, with a high bias of 10%–13% in the tropical upper troposphere that increases with increasing pressure [*Cooper et al.*, 2011].

[10] Ozonesonde measurements at Java (7°S, 111°E), Fiji (18°S, 178°E), Samoa (13°S, 171°W), Hilo (19°N, 155°W), San Cristobal (1°S, 89°W), and Natal (5°S, 35°W) are used here. These measurements are part of the Southern Hemisphere Additional Ozonesondes (SHADOZ, http://croc.gsfc.nasa.gov/shadoz/) network, a group of 16 ozonesonde sites in the southern tropics [*Thompson et al.*, 2003a, 2003b].

[11] In this study, we used the monthly mean Nino 3.4 index, based on sea surface temperatures (SST), to determine when El Niño and La Niña conditions occur. This index is available from the NOAA SST indices website (http://www.cpc.ncep.noaa.gov/data/indices/).

[12] Outgoing longwave radiation (OLR) serves as an indicator of deep convection. Data are provided by NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their website (http://www.esrl.noaa.gov/psd/).

3. Results

[13] Figure 1 shows the fraction of MLS soundings at the 215 hPa level that recorded low ozone concentrations (<20 ppb). These low concentrations are less than half of the mean tropical upper tropospheric ozone concentrations of 50 ppb. To our knowledge, this is the first map of low ozone event frequency. These low ozone events occur frequently in the tropical West Pacific and rarely occur elsewhere in the tropics. Figure 1 also shows a map of annual mean OLR for which lower values indicate regions with high clouds. A comparison of these plots shows some similar spatial features in tropical marine regions between the frequency of low ozone events and high clouds, suggesting that these events have a convective origin. Although central Africa and South America are also regions with strong convection, boundary layer ozone concentrations over land tend to be higher than those over ocean, and thus, deep convection does not create as many low ozone events. The fractions shown in Figure 1 agree within 12% with frequencies calculated from ozonesonde measurements over 1998–2008 from three

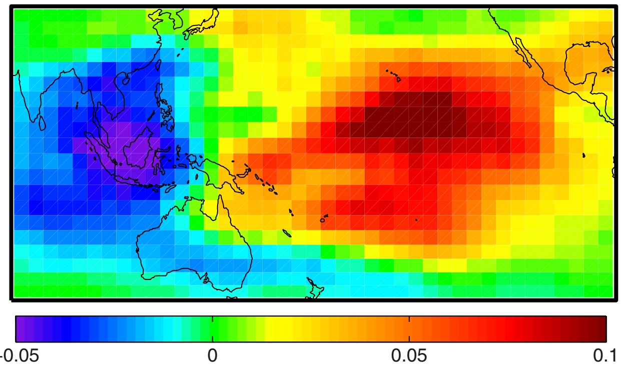

Figure 2. Difference in deseasonalized frequency of low ozone events between El Niño and La Niña conditions over years 2004–2012. El Niño (La Niña) conditions are defined using Nino 3.4 SST anomaly of greater than 0.4 (less than −0.4) K.

SHADOZ sites in the region (Samoa, Java, and Fiji) and with similar ozonesonde analyses in previous studies [*Solomon et al*., 2005]. Low ozone events occur exclusively within the pressure range of 100–300 hPa. The mean low ozone event frequency can vary by up to 20%.

[14] We repeated this analysis using data from the ACE-FTS and OSIRIS instruments and found features similar to those seen by MLS. Although ozonesonde comparisons indicate that MLS O_3 is more accurate at 215 hPa than at 147 hPa in the tropics [*Livesey et al*., 2012], we perform these comparisons at the 147 hPa level as errors in both ACE-FTS and OSIRIS increase with increasing pressure. Retrievals from all three satellites frequently show low ozone concentrations at 147 hPa over the tropical West Pacific (15°S–15°N, 100°E–180°E), with low ozone occurring in 11% of ~280,000 MLS, 11% of ~26,000 OSIRIS, and 12% of ~200 ACE-FTS retrievals. ACE-FTS and OSIRIS also observe low ozone concentrations less frequently in other regions, occurring in less than 5% of profiles elsewhere in the tropics. This is consistent with ozonesondes in Hilo, San Cristobal, and Natal, which also have O_3 concentrations less than 20 ppb in fewer than 5% of profiles. It is unlikely that artifacts in the retrievals would produce such consistent errors since each instrument measures radiation in a different spectral region.

[15] We use the chemical transport model GEOS-Chem v9-01-03 (see supporting information) to simulate low ozone events. The model is driven by assimilated meteorological fields from GEOS-5 for the years 2004–2010. We add Gaussian random noise ($\sigma = 20$ ppb) to the simulated ozone profiles to mimic MLS measurement precision. Without this added noise, the model simulates the spatial distribution of low ozone events but underestimates the frequency by up to 25%. The simulated frequency of low ozone events with added noise is displayed in Figure 1. The simulation reproduces the location of maximum frequency and has significant spatial agreement with MLS elsewhere ($r = 0.85$, $n = 75$). We focus our attention on the region of maximum frequency.

[16] *Lawrence et al*. [2003] found a strong sensitivity of upper tropospheric ozone to deep convection. We tested the hypothesis that these events are convectively driven by performing a GEOS-Chem simulation without convection. The frequency of low ozone events decreases relative to the standard simulation a few days after convection ceases, leading to a 15% increase in upper tropospheric O_3 concentrations. The simulated O_3 chemical production and loss rates change by less than 5%, which is insufficient to explain the upper tropospheric O_3 concentration changes over a few days. Thus, the decreasing frequency of low ozone events and resulting O_3 concentration increase can only be explained by the lack of transport of ozone poor air from the boundary layer.

[17] Previous analysis has attributed interannual variability in upper tropospheric ozone concentrations from MLS to dynamical variability due to the ENSO circulation [*Livesey et al*., 2012; *Oman et al*., 2013]. We further explore these low ozone events by separating the MLS retrievals into two groups representing either El Niño or La Niña conditions based upon the monthly mean Nino 3.4 index. Figure 2 shows the resulting change in low ozone event frequency. Low ozone events over the East Pacific occur almost twice as frequently during El Niño (increasing from 8% to 15% over 15°N–15°S, 180°W–100°W). This shift in the frequency of low ozone events mirrors the shift of the warm sea surface temperatures that occurs during El Niño events.

[18] Low ozone events occur more frequently over the East Pacific during El Niño conditions and more frequently over the West Pacific during La Niña. Table 1 shows that a shift similar to that seen by MLS is also evident in observations from the ACE-FTS and OSIRIS satellite instruments and in comparisons between ozonesondes in the East Pacific (Samoa) versus those in the West (Java). Uncertainties in the values given in Table 1 were estimated using the measurement precision for each instrument. The fraction of low ozone events found in all three satellites generally agrees within their uncertainties. The frequency change observed

Table 1. Low Ozone Fraction[a]

		East Fraction	East Number	West Fraction	West Number	Difference
MLS	El Niño	16 ± 4	66,000	11 ± 3	59,000	5 ± 5
	La Niña	8 ± 3	122,000	14 ± 3	92,000	−6 ± 4
OSIRIS	El Niño	26 ± 2	6000	14 ± 3	6,000	12 ± 4
	La Niña	14 ± 3	8000	16 ± 4	7,000	−2 ± 5
ACE-FTS	El Niño	26 ± 9	27	13 ± 8	16	13 ± 12
	La Niña	5 ± 3	41	19 ± 12	16	−14 ± 12
Ozonesondes	El Niño	28 ± 5	53	17 ± 5	91	11 ± 7
	La Niña	11 ± 3	109	31 ± 4	161	−20 ± 5

[a]Percentage of measurements at 145 hPa with ozone concentrations less than 20 ppb. Total number of observations in each region is also shown. West Pacific is bounded by 15°S–15°N, 70°E–140°E, and East Pacific is bounded by 15°S–15°N, 110°W–180°W. Ozonesonde measurements from Samoa (Java) are used for East (West) Pacific. Difference values are for East Pacific fraction minus West Pacific fraction.

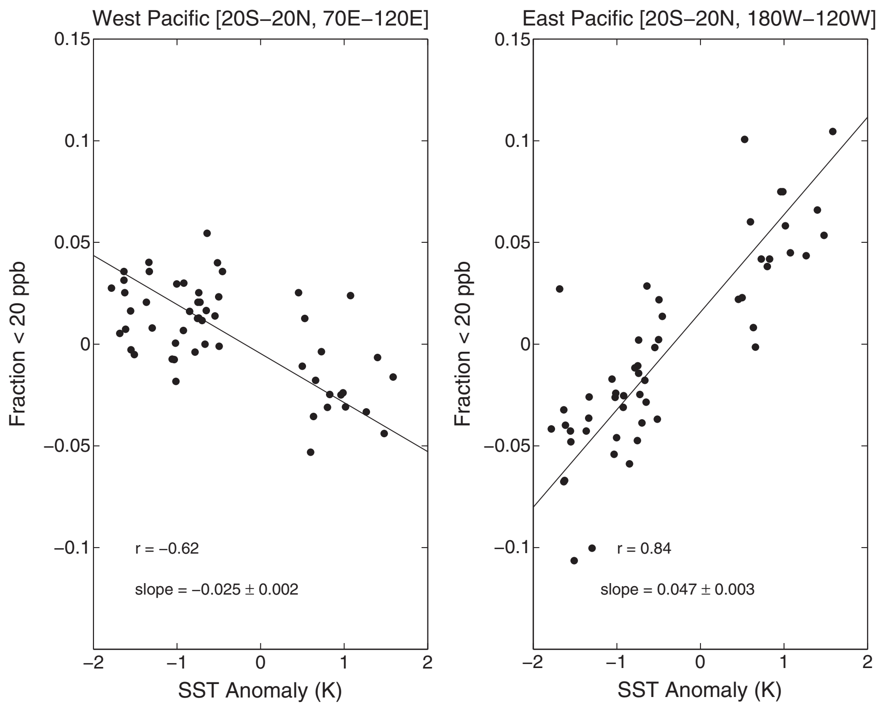

Figure 3. Scatterplot showing deseasonalized frequency of low ozone events versus Nino 3.4 sea surface temperature anomaly. Each dot represents a 3 month sliding average of MLS values. Only points during El Niño or La Niña conditions are shown. The slope and uncertainty of the best fit lines were found by a reduced major axis regression [*Miller and Kahn*, 1962].

by the satellite instruments due to ENSO is similar to that seen in the ozonesonde record, although positive biases in the satellite ozone retrievals may lead to fewer low ozone events, meeting the 20 ppb threshold. GEOS-Chem simulations for El Niño and La Niña years show an increase in low ozone event frequency over the central Pacific during El Niño but do not fully reproduce the observed decrease in frequency over the West Pacific since the GEOS-5 meteorological fields do not appropriately decrease convective activity in that region.

[19] In Figure 3, we test the hypothesis that larger temperature changes associated with stronger El Niño events have larger effects on convective frequency. Each dot represents a 3 month sliding average of the deseasonalized frequency of low ozone events in the MLS record over the East or West Pacific plotted against the Nino 3.4 SST anomaly. A statistically significant correlation (p values less than 10^{-7}) is evident between the value of the Nino 3.4 SST anomaly and the frequency of low ozone events on both sides of the Pacific. A large positive SST anomaly value indicates El Niño conditions, which leads convection to occur more frequently over the East Pacific and less frequently over the West. The slopes of the best fit lines indicate that the frequency of low ozone events is strongly related to the strength of the ENSO events.

[20] MJO signals are also visible in the MLS ozone record. The Hovmöller diagram in Figure 4 shows the low ozone event frequency as a function of time and longitude over the tropical Indian and Pacific oceans. We observe that regions with frequent low ozone events form over the Indian Ocean and travel eastward across the Pacific in a manner resembling similar diagrams made using outgoing longwave radiation measurements during the MJO [*Wong and Dessler*, 2007]. The eastward moving patterns occur with a period of 1–2 months, and we calculate an average propagation speed of 5 m/s. These values match those typically measured of the MJO. The GEOS-Chem simulation partially resolved eastward moving patterns of low ozone frequency, but further development of the meteorological fields appears needed.

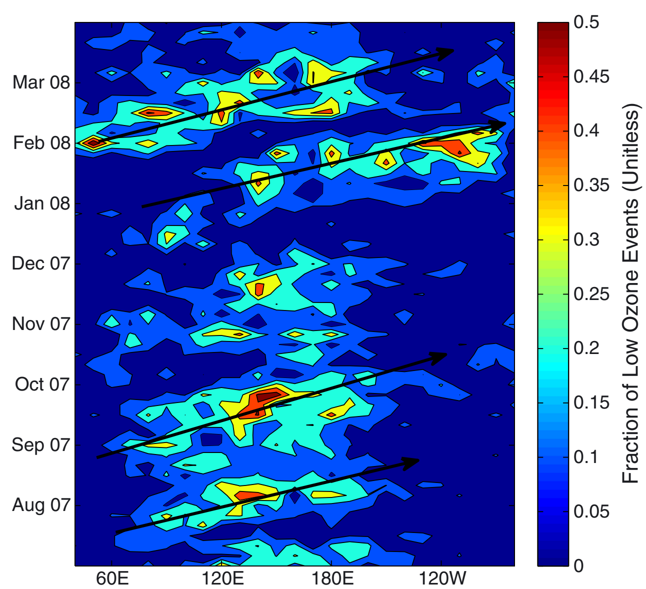

Figure 4. Hovmöller diagram displaying the low ozone event frequency from MLS over the Indian and Pacific Oceans for the period of August 2007 to March 2008. Areas of frequent low ozone events move eastward at ~5 m/s, in a repeating pattern with a period of 1–2 months, following the Madden-Julian Oscillation.

4. Conclusion

[21] Coherent spatial and temporal patterns of ozone concentrations less than 20 ppb in the tropical upper troposphere are observed for the first time with the MLS, ACE-FTS, and OSIRIS satellite instruments. These low ozone events occur most frequently in the convectively active region of the West Pacific warm pool with spatial patterns that resemble outgoing longwave radiation. The MLS, ACE-FTS, and OSIRIS satellite instruments observe low ozone events in 15%–25% of ozone profiles in this region, which agrees with values in the ozonesonde record. Both satellite and ozonesonde observations indicate shifts in low ozone events over the Pacific Ocean related to the ENSO circulation, with more frequent events over the central Pacific when the ocean beneath warms during El Niño conditions. On shorter time scales, regions with frequent low ozone events propagate eastward through the tropics with a period and a velocity typical to the MJO. Results from a state-of-the-science chemical transport model (GEOS-Chem) support the hypothesis that the events result from deep convection. The model was able to simulate the structure of the most frequent low ozone events but only partially reproduced the westward shift in low ozone event frequency during La Niña. Future effort to reproduce these features will require developments to convective parameterizations. These observations provide a sensitive test of the ability of global models to represent the coupling of chemical and dynamical processes of relevance to climate.

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