Evaluation of lightning-induced tropospheric ozone enhancements observed by ozone lidar and simulated by WRF/Chem

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HIGHLIGHTS

• Lidar measures ozone profiles at Huntsville, AL on July 14, 18 and 27, 2011.
• Lightning-NOx emission (LNOx) is estimated based on NLDN flash measurements.
• LNOx emission applies to WRF/Chem for simulating lightning influence.
• Lidar-measured ozone variation is compared to model.
• Upwind lightning resulted in ozone enhancements at Huntsville on July 14 and 27.

ABSTRACT

High spatial- and temporal-resolution ozone lidar profiles, in conjunction with ozonesonde and satellite observations, are well suited to characterize short-term ozone variations due to different physical and chemical processes, such as the impact of lightning-generated NOx (LNOx) on tropospheric ozone. This work presents the hourly variation of tropospheric-ozone profiles measured by an ozone lidar at the University of Alabama in Huntsville, on July 14, 18, and 27, 2011. These ozone lidar data are compared with two WRF/Chem simulations, one with lightning NO (LNO) emissions and the other without. On July 14, 2011, the ozone lidar observed an ozone laminar structure with elevated ozone concentrations of 65–80 ppbv below 2 km, low ozone (50–65) ppbv between 2 and 5 km, and high ozone up to 165 ppbv between 5 and 12 km AGL. WRF/Chem simulations, in conjunction with backward trajectory analysis, suggest that lightning events occurring within upwind regions resulted in an ozone enhancement of 28 ppbv at 7.5 km AGL over Huntsville. On July 27, LNO emissions were transported to Huntsville from upwind and account for 75% of NOx and an 8.3 ppbv of ozone enhancement at ~10 km; the model overestimates ozone between 2.5 and 5 km AGL.

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1. Introduction

Lightning is a particularly significant NOx source in the middle and upper troposphere where it affects tropospheric chemistry and ozone (Ridley et al., 1996; Huntrieser et al., 1998; Pickering et al., 1998; Zhang et al., 2000, 2003; Bond et al., 2001, 2002; Hudman et al., 2007; Allen et al., 2010, 2012). In-situ photochemistry and dynamic processes govern the ozone distribution in the troposphere where ozone is a precursor of the hydroxyl radical that reacts with most trace gas species. Ozone is an important greenhouse gas, influencing the climate by its radiative forcing (Lelieveld and Dentener, 2000; Lacis et al., 1990). More than 80% of summertime upper tropospheric NOx above the eastern United States is produced by lightning, assuming a lightning NOx emission rate of 250 mol per flash (Cooper et al., 2009). Li et al. (2005) first suggested a summertime ozone maximum within the semi-permanent
upper-tropospheric anticyclone above eastern North America, and this ozone enhancement is largely associated with lightning-produced NOx (LNO). Based on ozonesonde data provided by the INTEX Ozone Network Study (IONS) experiments in summer 2004 and 2006, Cooper et al. (2006, 2007) confirmed that the upper-tropospheric ozone maximum is a recurring summertime feature, and estimated that LNO emissions led to the production of 25–30 ppbv of ozone at 250 hPa above the southern United States during August 2006. Wang et al. (2013) lessened the upper tropospheric low bias in ozone by ~20 ppbv above the southeastern and eastern U.S.A after they included lightning-produced NO emissions into the version-4 Community Multiscale Air Quality Modeling System (CMAQ).

High resolution and continuous observational capability make lidars ideal instruments to capture rapid variations in ozone distribution (Kuang et al., 2011a, 2012). This paper evaluates the tropospheric ozone variability associated with lightning events as measured by the Rocket-city O3 Quality Evaluation in the Troposphere (RO3QET) lidar in Huntsville, AL, and simulated by the Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-Chem). It is the first study to use back trajectories and an air quality model to relate ozone laminae in lidar profiles to NO with a lightning source.

2. RO3QET ozone differential absorption lidar (DIAL) at UAHuntsville

The RO3QET DIAL, one of the Tropospheric Ozone Lidar Network (TOLNet: http://www-air.larc.nasa.gov/missions/TOLNet/index.html) stations, is located on the campus of University of Alabama in Huntsville (UAHuntsville) (34.72N, 86.65W) at 206 m ASL. The system operates with two wavelengths at 285 and 291 nm and employs two receivers with 10 and 40-cm primaries. Ozone is retrieved from 0.5 to ~12 km AGL during both daytime and nighttime. Because the backscattered laser signal decays quickly with range, the vertical resolution of the DIAL retrievals is designed to vary with altitude (Godin et al., 1999) so as to obtain a balance between vertical resolution and statistical errors. In this manner, the effective vertical resolution for DIAL retrievals changes from ~150 m in the PBL to ~750 m in the upper troposphere. Both analysis and validation against ozonesonde measurements demonstrate that this lidar measures ozone with a precision generally better than ±10% in the lower troposphere and better than ±20% in the upper troposphere, at a typical temporal resolution of 10 min (Kuang et al., 2011b, 2013). However, the longer integration time (1-hr) used in this paper is expected to improve the retrieval precision to ±4% in the lower troposphere and ±8% in the upper troposphere. Sub-hourly variation of tropospheric ozone profiles was measured on July 14, 18, and 27, 2011.

3. WRF-Chem model

Simulations were conducted using the Weather Research and Forecasting model with Advanced Research WRF (ARW) core V3.3.1 with online chemistry (Skamarock et al., 2005; Grell et al., 2005). The model was initialized on June 27, 2011 1200 UTC and ran through 0000 UTC August 2, 2011. The model domain covers the CONUS with 170 x 103 grid cells and a horizontal resolution of 36 km. There are 34 vertical layers ranging from the surface to ~50 mb, where the vertical resolution in the upper troposphere is ~20–50 mb, depending on the level. Meteorological initial and boundary conditions were obtained from the National Centers for Environmental Prediction (NCEP) reanalysis (Kalnay et al., 1996). Chemical initial and boundary conditions were generated at 6-h time intervals by the MOZART-4 global chemical transport model (Emmons et al., 2010). Objective analysis (OBGRID) nudging was used to constrain the model by incorporating information from observations. For this analysis, high resolution upper level and surface observations were combined with the global analysis fields (Yegorova et al., 2011). In addition, Four-Dimensional Data Assimilation (FDDA) nudging was used. Temperature, water vapor, and winds were nudged towards analysis fields. Table 1 contains a list of the major physical, dynamical, and chemical options chosen for the simulation.

Anthropogenic emissions were processed with the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System (Houyoux and Vukovich, 1999) using an EPA-projected 2012 emissions inventory. Biomass burning emissions were obtained from the Fire Inventory from NCAR version 1.0 (FINNv1; Wiedinmyer et al., 2011) and were vertically distributed based on a plume rise algorithm within the WRF/chem modeling system. Biogenic emissions were calculated within WRF/Chem using the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.0; Guenther et al., 2012).

For the period of June 27 to August 2, 2011, we conducted two WRF-Chem model runs, one with lightning-produced NO emissions (LNO) (hereafter referred to as “LGT” run) and the other without (hereafter referred to as “CNTRL” run).

4. LNO emissions

National Lightning Detection Network™ (NLDN; Cummins and Murphy, 2009) Cloud-to-Ground (CG) flash data for July 2011 from the Global Hydrology and Climate Center (GHCC) lightning team (http://thunder.mscf.nasa.gov/), divided by 0.93 for a rough mean of the 90–95% NLDN CG detection efficiency, is mapped onto the 36 km by 36 km WRF/Chem model domain on an hourly basis. Fig. 1a shows the accumulated CG flashes over the continental U.S. (CONUS) during the entire month of July. The Intra-Cloud (IC) flash rates are produced by applying the climatological IC-to-CG ratios for July 2011 (Boccippio et al., 2001) to the detection-efficiency adjusted NLDN CG flashes. Assuming that IC flashes are as energetic as CG flashes (DeCaria et al., 2005; Ott et al., 2007; Fehr et al., 2004) and that each flash produces approximately 500 mol of NO on average (Ott et al., 2010), we calculate the LNO production for each flash, and vertically distribute the LNO amount following an LNO emission profile (Fig. 1b) derived by convolving pressure with the warm season mean segment altitude distribution from the Northern Alabama Lightning Mapping Array data (Koshak et al.,

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<tr>
<td>Chemistry option</td>
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<td>MOSAIC with 8 sectional aerosol bins</td>
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<td>Photolysis option</td>
<td>Fast-J</td>
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* Details about the configuration options listed, and references for the physical parameterizations used in each module can be found in Grell et al. (2005) and at http://ruc.fsl.noaa.gov/wrf/WG11.1;
The LNO emission profile used in this study has been adjusted to reflect LNO dependence on atmospheric pressure (under the same conditions, higher pressure yields more LNO production) (Wang et al., 1998). We place the LNO emissions at the time and location of the NLDN CG flashes, to ensure that LNO emissions are placed at the exact time and location as was observed by NLDN (Kaynak et al., 2008; Smith and Muller, 2010; Wang et al., 2013).

5. Results and analyses

5.1. Modeled lightning contribution to NOx

The difference in NOx concentration between LGT and CNTRL, dividing by LGT-simulated NOx, is calculated to represent the fractional lightning contribution to the atmospheric NOx (Fig. 2a). Averaged over the July 2011 period, lightning contributes more than 60% of the calculated NOx between 5 and 10.5 km altitudes over the CONUS domain (black dot-dashed). In comparison to the CONUS, Huntsville (black) has a wider range of altitudes (3.4–12.8 km AGL) where lightning contributes more than 60% of NOx. The lightning contribution increases to more than 70% between 3.9 and 10.3 km with a peak of 80% between 5 and 6 km.

Model output for the Huntsville location (HSV), averaged over three lidar observation periods: 10 to 17 Local Time (LT), July 14, 10 to 17 LT, July 18, and 11 to 17 LT, July 27, are also plotted in Fig. 2. Lightning contributes 80% to the NOx concentrations at 7.5 km on July 14, and 75% around 10 km on July 27, both exceeding the Huntsville monthly mean values at the corresponding altitudes (Fig. 2a). We will discuss the lightning sources that resulted in these NOx enhancements in 5.3.

5.2. Modeled O3 enhancement due to lightning

The ozone differences between LGT and CNTRL (as shown in Fig. 2b), indicate the ozone enhancements due to lightning. Averaged for July 2011, lightning enhances tropospheric ozone concentration throughout the CONUS domain with a maximum enhancement of 4.5 ppbv (7%) at ~6.7 km AGL (black dot-dashed). The Huntsville monthly-mean O3 enhancement due to lightning (black) is about 1.7–3.6 times larger than the O3 increase over the CONUS, with a maximum enhancement of 12.4 ppbv (18%) at ~7.3 km AGL.

Fig. 1. (a) Total Cloud-to-ground (CG) flashes observed by NLDN during July 2011, adjusted with a detection efficiency of ~93%. (b) Vertical distribution of LNO emission at 0.1 km resolution, derived from the vertical distribution of warm-season mean segment altitude distributions from the Northern Alabama Lightning Mapping Array data.

Fig. 2. (a) WRF/Chem-simulated lightning contribution to NOx; (b) WRF/Chem-simulated ozone enhancement due to lightning. “LT” stands for local time.
The red solid line in Fig. 2b represents model-simulated ozone enhancement averaged between 10 and 17 LT on July 14 during which the lidar observation was made. A maximum ozone enhancement of 28 ppbv occurs at 7.5 km AGL, which greatly exceeds the Huntsville monthly-mean ozone enhancement of 13 ppbv at this altitude. Together with the 80% lightning contribution to atmospheric NOx shown in Fig. 2a, this ozone enhancement of 28 ppbv suggests a large lightning influence on upper tropospheric ozone above Huntsville on July 14. Further discussion is in Section 5.3.

5.3 Modeled O3 versus lidar measurements

The original lidar measurements have a 10 min temporal resolution and a vertical resolution ranging from 150 m in the planetary boundary layer (PBL) to 750 m in the upper troposphere. To compare with model output, we interpolated the original lidar data onto the 34 model layers and averaged on an hourly basis. Fig. 3 compares modeled (CNTRL and LGT) and lidar-observed O3 variations on July 14, 18, and 27, 2011, respectively. The bottom panels in Fig. 3a show these spatially-interpolated hourly-mean lidar values.
on July 14, 18, and 27, respectively. The temporally-averaged lidar profiles are plotted in Fig. 3b.

On July 14, 2011, the lidar observed elevated PBL ozone (65–80 ppbv below 2 km altitude) (Fig. 3a, bottom left panel). There exists a low-ozone layer of 50–65 ppbv between 2 and 5 km which keeps decreasing gradually with time and an enhanced ozone layer above 5 km. The LGT run well-captures this lidar-observed ozone laminar structure of high O3 in the PBL, low O3 between 2 and 5 km, and high O3 again above 5 km (Fig. 3a, middle left panel). The LGT-simulated ozone agrees well with lidar observation above 5 km, although it slightly overestimates between 1 and 3 km (Fig. 3b). Starting from 5 km, the CNTRL run significantly underestimates O3. Both runs underestimate ozone above 9 km, however, with lightning inputs, the LGT run has a smaller negative bias than the CNTRL. Overall, the LGT performs better than CNTRL, suggesting the significant influence of lightning on July 14.

Fig. 4. (a) Backward trajectories calculated using WRF 36 km wind fields, starting from 18 UTC, July 14, 2011, at Huntsville, AL, backward for 24 h. Backward trajectories at 9000, 8000, 7000, 6000, 5000, 4500, 4000, 3500, and 3000 m AGL. (b) A closer view of the trajectories at 4500, 4000, 3500, and 3000 m. (c) 24-h precipitation ending at 0700 E.S.T., July 13, 2011 (Daily weather map: http://www.wpc.ncep.noaa.gov/dailywxmap/index_20110713.html). (d) 24-h precipitation ending at 0700 E.S.T., July 14, 2011 (Daily weather map: http://www.wpc.ncep.noaa.gov/dailywxmap/index_20110714.html). (e) Total Cloud-to-ground (CG) flashes observed by NLDN during the 24-h period ending at 0700 E.S.T., July 13, 2011. (f) Total Cloud-to-ground (CG) flashes observed by NLDN during the 24-h period ending at 0700 E.S.T., July 14, 2011.
and west and southwest of Huntsville within the 24-hour period ending at 1200 UTC, July 14, respectively. NLDN observed robust lightning activities in these areas (Fig. 4e and f). Using the WRF-simulated wind field, we calculated 24-hour backward trajectories starting from 1800 UTC (1 pm LT), July 14, at eight altitudes ranging from 3000 m to 9000 m AGL above Huntsville (Fig. 4a and b). In addition, backward trajectories calculated from HYbrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) using several sets of independent meteorological data (e.g., NAM 12 km, NCEP/NCAR Reanalysis, GDAS) all yield similar results. The heavy precipitation in southern Kentucky, Missouri, and Illinois within the 24-hour period ending at 1100 UTC, July 13 correspond nicely with the 7000 m, 8000 m, and 9000 m trajectories, as well as the very large ozone enhancements simulated at those altitudes on July 14 by the LGT run (Fig. 3b). It looks like storms occurring within the 24-hour period ending at 1100 UTC, July 13 led to LNOx-enhanced ozone at those levels, which was then transported to HSV region. The trajectories starting at 3500 m (red), 4000 m (green), and 4500 m (dark pink) AGL passed through the region west and southwest of Huntsville where a lot of precipitation occurred within the 24-hour period ending at 1100 UTC, July 14 (Fig. 4d). Therefore, the lidar-observed ozone enhancement at Huntsville on July 14 was contributed by lightning events taking place in southern Kentucky, Missouri, Illinois, and west and southwest of Huntsville within the previous two days (July 12 and July 13).

On July 18, the lightning-influenced ozone within the 2–5.5 km layer is greater than the CONUS monthly mean but less than Huntsville monthly mean (Fig. 2b). Between 2 and 5.5 km, the LGT run enhances ozone by 6–7 ppbv and improves agreement with the lidar observations (Fig. 3b). Both runs underestimated surface O3 by about 13 ppbv. Above 11 km, the lidar measures 125 ppbv of ozone whereas the model estimate is only 70 ppbv.

On July 27, both runs overestimate ozone by up to 22–30 ppbv between 2.5 and 5 km (Fig. 3a and b). The model simulates extremely dry conditions (in comparison to July 14 and 18) within this vertical range on this day, but there's currently no evidence to show any potential linkage between this model-simulated dry air condition and overestimated ozone. The LGT run agrees well with the lidar observations around 10 km, by simulating 8.3 ppbv more ozone than the CNTRL (although it’s not a very significant improvement considering the big measurement uncertainty there. Averaged over 11–17 LT on July 27, 75% of the NOx (0.134 out of 0.181 ppbv) at 10 km is from lightning contribution (Fig. 2a). Because NLDN flash data shows no lightning events occurred at Huntsville on July 27, we calculated back trajectories using WRF-simulated wind fields to find upwind lightning sources that account for this enhanced NOx. At 00UTC, July 27, the backward trajectories pass through the southwest edge of a thunderstorm in northern Kentucky and southern Ohio, where NLDN data reveals the occurrence of lightning events. Comparison between the two model runs shows that this lightning event adds 0.76 ppbv of NO around 10 km altitude there (Fig. 5b). Sixteen hours later, this NO-enhanced air mass was transported to Huntsville and elevated the 10-km NOx from 0.047 ppbv to 0.181 ppbv.

6. Summary and conclusion

This is the first study to employ high spatial- and temporal-resolution ozone-lidar profiles to evaluate the impact of lightning on tropospheric ozone. We investigate the impact of lightning on tropospheric ozone enhancements observed by lidar at Huntsville, AL, on July 14, 18, and 27, 2011 using two WRF/Chem simulations, one with LNO emissions and the other without. On July 14, 2011, the ozone lidar observed ozone laminar structure with enhanced ozone concentrations in the boundary layer and above 5 km AGL, which is also captured by the WRF/Chem simulation that includes LNO emission. Backward trajectory analysis reveals that lightning events occurring west/southwest of Huntsville as well as storms occurring in southern Kentucky, Missouri, and Illinois account for the HSV ozone enhancement simulated by LGT run and observed by the lidar. On July 27, both CNTRL and LGT runs overestimate ozone by 22–30 ppbv between 2.5 and 5 km above Huntsville, with extremely dry conditions simulated within the same vertical range. Lightning occurring near the southwest edge of a thunderstorm in northern Kentucky and southern Ohio at 0000 UTC, July 27 adds about 0.76 ppbv of NO around 10 km altitude in this region. Back trajectories indicate this air mass with elevated NO was transported to Huntsville after 16 h, increasing NOx by 0.134 ppbv, and resulting in an ozone enhancement of 8.3 ppbv at 10 km AGL. For July 18, lightning has very small influence on the HSV ozone concentration.

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References


