Lightning NO\textsubscript{x} in the 29–30 May 2012 Deep Convective Clouds and Chemistry (DC3) Severe Storm and Its Downwind Chemical Consequences

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Abstract A cloud-resolved storm and chemistry simulation of a severe convective system in Oklahoma constrained by anvil aircraft observations of NO\textsubscript{x} was used to estimate the mean production of NO\textsubscript{x} per flash in this storm. An upward ice flux scheme was used to parameterize flash rates in the model. Model lightning was also constrained by observed lightning flash types and the altitude distribution of flash channel segments. The best estimate of mean NO\textsubscript{x} production by lightning in this storm was 80–110 mol per flash, which is smaller than many other literature estimates. This result is likely due to the storm having been a high flash rate event in which flash extents were relatively small. Over the evolution of this storm a moderate negative correlation was found between the total flash rate and flash extent and energy per flash. A longer-term simulation at 36-km horizontal resolution with parameterized convection was used to simulate the downwind transport and chemistry of the anvil outflow from the same storm. Convective transport of low-ozone air from the boundary layer decreased ozone in the anvil outflow by up to 20–40 ppbv compared with the initial conditions, which contained stratospheric ozone. Photochemical ozone production in the lightning-NO\textsubscript{x} enhanced convective plume proceeded at a rate of 10–11 ppbv per day in the 9–11 km outflow layer over the 24-hr period of downwind transport to the Southern Appalachians. Photochemical production plays a large role in the restoration of upper tropospheric ozone following deep convection.

Plain Language Summary Nitrogen oxides are important precursors for tropospheric ozone, an important greenhouse gas. The global amount of nitrogen oxides produced by lightning remains highly uncertain, primarily because of uncertainty in the amount produced per flash. In this paper we use an approach that involves cloud-resolved modeling with chemistry, constrained by observed flash rates and aircraft measurements of nitrogen oxides, to make an estimate of the mean production per flash for an observed severe storm over Oklahoma. We estimate that the mean production rate was 80–110 mol per flash for this flash rate storm, which is at the lower end of the range reported in the literature. We use a regional model to follow the outflow of the Oklahoma storm downwind to estimate the amount of ozone produced in the upper tropospheric outflow plume by the lightning-generated nitrogen oxides. Our estimate is in the range found in previous studies, and we note that the ozone production by photochemistry is an important process in restoring upper tropospheric ozone following storms that lofted low values of ozone from the layer of air near the surface.

1. Introduction

As one of the largest natural sources of nitrogen oxides (NO, nitric oxide (NO), nitrogen dioxide (NO\textsubscript{2})), lightning accounts for roughly 5–3 Tg (N) yr\textsuperscript{-1}, or 10%–15%, of the total NO\textsubscript{x} budget (Schumann & Huntrieser, 2007). While the lower troposphere is mainly dominated by surface emissions, especially those due to human activity (e.g., Zhang et al., 2003), lightning is the largest source of NO\textsubscript{x} in the upper troposphere, where the majority of lightning NO\textsubscript{x} (LNO\textsubscript{x}) is present in cloud outflow above an altitude of 7 km (Martin et al., 2007; Ott et al., 2010; Pickering et al., 1998). In this portion of the atmosphere, NO\textsubscript{x} well downwind of thunderstorms has a longer lifetime than in the boundary layer and a large influence on ozone (O\textsubscript{3}) (e.g., D. Allen et al., 2010), a significant greenhouse gas (Lacis et al., 1990), having a positive radiative forcing on the climate. Following a convective event, enhancements in O\textsubscript{3} can be found downwind within the thunderstorm outflow (Apel...
Many factors add uncertainty to LNO\textsubscript{x} production, including variation in the type, length, energy, peak current, and number of lightning flashes (Price et al., 1997), as well as the location and strength of the convection. Although other natural and anthropogenic NO\textsubscript{x} sources may have uncertainty ranges that are similar to that associated with lightning, the potential feedback mechanisms between lightning and surface temperatures makes narrowing the uncertainty range of LNO\textsubscript{x} critical (Schumann & Huntrieser, 2007). In addition, a better understanding of the global LNO\textsubscript{x} budget would be beneficial for properly modeling variations and trends in NO\textsubscript{x} and O\textsubscript{3} and in analyzing the influence of other NO\textsubscript{x} sources. Therefore, properly simulating lightning flash rates and LNO\textsubscript{x} production has the potential to enhance our knowledge of the role of LNO\textsubscript{x} in global climate-chemistry models (e.g., Banerjee et al., 2014; Liaskos et al., 2015) and regional air quality models (e.g., D. J. Allen et al., 2012; Kang et al., 2019). Past and future model simulations are valuable for furthering our understanding of the potential impacts of climate change on convection and the resulting lightning activity (Finney et al., 2018; Magi, 2015; Murray, 2018; Petersen & Rutledge, 2001; Price, 2009; Reeve & Touni, 1999; Romps et al., 2014; Williams, 2005).

The Deep Convective Clouds and Chemistry (DC3) field campaign (Barth et al., 2015) made simultaneous observations of the microphysics, dynamics, chemistry, and lightning within convective systems, as well as post-convection observations of the downwind plume. The availability of this large, cohesive data set provides the necessary surface and upper air measurements required for model simulations of electrical and chemical relationships within thunderstorms.

Cummings et al. (2024) tested numerous flash rate parameterization schemes (FRPSs) in Weather Research and Forecasting model simulations of the 29–30 May 2012 DC3 severe storm that occurred in Oklahoma. The FRPS that yielded the best prediction of total flashes over the period from storm initiation through the end of aircraft sampling of the storm system was the upward cloud ice flux scheme of Finney et al. (2014). We use this scheme in the cloud-resolved WRF-Chem simulations for that same storm to address the following research questions:

1. What is the best estimate of the mean LNO\textsubscript{x} production efficiency (PE) in terms of moles NO\textsubscript{x} per flash?
2. How do observed versus default model vertical distributions of flash channel lengths and intracloud to cloud-to-ground (IC:CG) ratios affect the model-simulated LNO\textsubscript{x} and other trace gases in the anvil outflow?
3. Does LNO\textsubscript{x} production per flash in this high flash rate Oklahoma storm vary from other midlatitude storms?
4. How do flash extent and flash energy relate to flash rate for the Oklahoma severe supercell system in comparison to other observed storms?
5. Is there a relationship between flash rate and LNO\textsubscript{x} production per flash?

One of the primary objectives of this research is to use the 29–30 May storm simulation, constrained by the observed lightning mapping array (LMA) flash rate and characteristics and by aircraft observations of NO\textsubscript{x}, to realize a best estimate of the mean LNO\textsubscript{x} production per flash for this storm. Model experiments designed to make this estimate are discussed in Section 4.1 and results are presented in Section 5.1. Pollack et al. (2016) made estimates of LNO\textsubscript{x} production for this storm using volume- and flux-based methods that employed the aircraft observations obtained in the anvil. Significant uncertainties exist in these estimates. Our modeling-based estimate provides an entirely different approach (although using the same observational data set), yielding an important confirmation of the Pollack et al. work. Comparison of our results with those of Pollack et al. is presented in Section 6.1.

We also run WRF-Chem at coarser horizontal resolution with parameterized convection to examine the evolution of the upper tropospheric outflow from the same Oklahoma severe storm over 24 hr of downwind transport. The primary questions posed for this simulation are:

1. By how much does the upper tropospheric O\textsubscript{3} in convective outflow change during transport
2. How much of this change can be attributed to photochemical production that has been enhanced by LNO\textsubscript{x}?
Section 2 of the paper summarizes previous model simulations of the 29–30 May DC3 storm and previous model estimates of LNOx PE and downwind O₃ production. Section 3 describes the DC3 experiment and presents the lightning and NOₓ observations. Section 4 describes the WRF-Chem model set up for the cloud-resolved storm simulation and the longer-term regional simulation with parameterized convection. Section 5 presents the model results for both simulations, and Section 6 discusses these results in relation to prior work. Section 7 offers some concluding remarks.

2. Previous Model Results

2.1. DC3 Storms

The 29–30 May 2012 storm has been simulated at cloud-resolving resolution by three previous investigators. Bela et al. (2016) presented results of a cloud-resolved simulation of this storm and its chemistry with emphasis on examination of the wet scavenging process. In particular, comparisons of aircraft data with various assumptions of the amount of retention of dissolved trace gases during freezing were performed. The soluble species most relevant to LNOₓ calculations is HNO₃, and Bela et al. (2016) found that HNO₃ was not sensitive to values tested for ice retention fractions. Using the same WRF-Chem configuration, Cummings et al. (2024) tested 18 FRPSs, finding that FRPSs based on upward cloud ice flux (Finney et al., 2014) and updraft volume (Deierling & Petersen, 2008) provided the best representation of lightning activity in the early and later stages of storm simulation, respectively. Li et al. (2017) examined convective transport of trace gases in cloud-resolved simulations of this severe storm, as well as two additional DC3 storms (an air mass thunderstorm and a mesoscale convective system (MCS)). They found through analyses of the vertical flux divergence in these three storms that the deep convective vertical transport per unit area was greatest in the 29–30 May storm. Bela et al. (2018) studied the wet scavenging process for soluble gases in these same three storms through analysis of the aircraft observations and the model experiments, finding that liquid-phase microphysical scavenging was the dominant process for CH₃O and H₂O₂ removal. Li et al. (2018) simulated the 29–30 May severe storm and the MCS at 36- and 12-km horizontal resolution, finding that subgrid-scale convective transport was more significant relative to that at grid scale in the 29–30 May severe storm than in the MCS case. Li et al. (2019) added ice retention factors to the subgrid convection and improved cloud to rain conversion factors for below freezing temperatures in simulations for the 29–30 May case.

2.2. Previous LNOₓ Simulations

Cloud-resolved simulations have been conducted for observed storms in previous field projects with various models to estimate NO production per flash and the Pₑ/Pₑₑ ratio, where Pₑₑ and Pₑₑₑ are the mean NO production per IC and CG flash, respectively. For all simulated storms, the model LNOₓ was compared against aircraft NOₓ observations in the anvil to deduce Pₑₑ and Pₑₑₑ. DeCaria et al. (2000, 2005) used realistic distributions of lightning channels and observed IC and CG flash rates. The flash channel vertical modes, as well as NOₓ production, followed Gaussian distributions. This general technique has also been used by Ott et al. (2007, 2010) in simulating a series of midlatitude and subtropical thunderstorms. The assumptions that Pₑₑₑ was roughly 460 mol flash⁻¹ and Pₑₑ was 75%–100% of Pₑₑₑ (DeCaria et al., 2005) provided the best comparison to the column NOₓ mass computed from aircraft observations for the 12 July 1996 Stratosphere-Troposphere Experiment: Radiation, Aerosols and Ozone (STERAO-A) storm. Fehr et al. (2004) and Ott et al. (2007) both studied the production of LNOₓ in the midlatitude 21 February 2011 Lightning Nitrogen Oxides Project (EULINOX) storm using different models. Similar results for Pₑₑₑ were obtained and both simulations showed an IC flash produced more LNOₓ than a CG flash. Ott et al. (2010) summarized the LNOₓ production results from five 3-D midlatitude and subtropical storm simulations. Mean production per CG flash over the five storms was 500 mol flash⁻¹ with a mean Pₑₑₑ/Pₑₑₑₑ ratio of 0.94. Cummings et al. (2013) performed cloud chemistry simulations for a Hector thunderstorm observed on 16 November 2005 during the SCOUT-O3/ACTIVE campaigns based in Darwin, Australia, with the primary objective of estimating the average NO production per lightning flash in this unique storm type which occurred in a tropical island environment. Lightning NO production scenarios of 500–600 mol flash⁻¹ for both CG and IC flashes yielded anvil NOₓ mixing ratios that compared well with aircraft observations.

The STERAO-A storm simulation by DeCaria et al. (2005) indicated that additional ozone production attributable to lightning NO within the storm cloud during the lifetime of the storm was very small (≤ 2 ppbv). However,
simulation of the photochemistry over the 24 hr following the storm showed that an additional 10 ppbv of ozone production in the upper troposphere can be attributed to lightning NO production. Convective transport of HOx precursors led to the generation of a HOx plume, which substantially aided the downstream ozone production. Ott et al. (2007) simulated the 21 July 1998 EULINOX thunderstorm. During the storm, the inclusion of LNOx in the model combined with convectively-transported boundary layer NOx from the Munich, Germany region resulted in sufficiently large NOx mixing ratios to cause a small loss of ozone (on average less than 4 ppbv) at all model levels. Simulations of the chemical environment in the 24 hr following the storm showed on average a small increase in the net production of ozone at most levels resulting from LNOx, maximizing at approximately 5 ppbv per day at 5.5 km. Between 8 and 10.5 km, LNOx caused decreased net ozone production. Ren et al. (2008) found that net tropospheric ozone production proceeded at a median rate of 11 ppbv per day above 9 km in the Intercontinental Transport Experiment (INTEX-A) in which the effects of frequent deep convection over the United States dominated the upper troposphere. Apel et al. (2012) noted from a box model calculation a net ozone increase of 10 ppbv over a few hours following observed convection with lightning over Canada in the Arctic Research of the Composition of the Troposphere from Aircraft and Satellite (ARCTAS) experiment. Apel et al. (2015) performed box modeling of the chemistry downwind of two DC3 storms in northeast Colorado on 22 June 2012. The northern storm ingested fresh biomass burning smoke, and the southern storm was affected by more aged biomass burning emissions. The model predicted substantial downwind ozone production in the UT for both storms. The southern storm was predicted to produce more ozone over 2 days (14 ppbv) than the northern storm (11 ppbv) despite having lower VOC OH reactivity. Sensitivity tests showed that this was principally due to more NOx being present in the southern storm outflow because of LNOx. Brune et al. (2018) found general agreement between observed and modeled OH and HO2 in the outflow of the 21 June 2012 DC3 MCS. In this study the DC-8 made multiple passes through the outflow as it moved downwind. Box model calculations yielded a 13 ppbv increase in ozone over 5 hr, similar to the observed 14 ppbv increase. This rate of increase is larger than others in the literature, perhaps because for a portion of the 5 hr the outflow was in cirrus cloud, in which photolysis rates may have been larger than clear-sky values due to multiple scattering.

3. DC3 Observations

3.1. Experiment Design

The DC3 field campaign provided a multidisciplinary approach to study different types of convection (e.g., midlatitude airmass, multicell, and supercell thunderstorms), their convective transport of trace gases (e.g., anthropogenic, biogenic, and wildfire emissions), and associated lightning occurrence and NOx production Barth et al. (2015). The DC3 operations center was in Salina, Kansas, which allowed reasonable flight times for research aircraft to arrive at any of the three designated sampling regions in northeast Colorado, central Oklahoma to west Texas, and northern Alabama. The research aircraft used in the experiment were the NASA DC-8, the National Science Foundation (NSF)/NCAR GV, and the DLR Falcon. Each aircraft was outfitted for measuring a variety of trace gases, aerosols, and meteorological variables. The three domains were selected based on the available instrumentation (radar, ground-based LMAs and sounding units) and type of convection associated with the region. Storm sampling took place from mid-May through June 2012. Most flights focused on active convection, but in a few cases the upper tropospheric outflow was sampled as much as 24 hr downwind. Additional details regarding the DC3 field campaign can be found in Barth et al. (2015).

The analyses in this paper focus on the 29–30 May 2012 storm which developed in northern Oklahoma. Lightning coverage was provided by the Oklahoma LMA (Bruning, 2014). The total lightning flash detection efficiency provided by the 11 stations in this LMA network is >90% at a 200 km radius (Chmielewski & Bruning, 2016). In addition to the National Weather Service WSR-88D Doppler radar network data (NCAR, 2012b), dual-Doppler radar coverage was performed by the NOAA National Severe Storm Laboratory’s X-band polarimetric (NOXP) and University of Oklahoma C-band Shared Mobile Atmospheric Research and Teaching (Shared Mobile Atmospheric Research and Teaching) radars. The NASA DC-8 and NSF/NCAR GV provided flight data (NASA, 2016) that included both boundary layer inflow and upper tropospheric anvil outflow sampling. On the DC-8 CO was measured by the NASA Langley Differential Absorption CO Measurement (DACOM) system, O3 and NO were observed with the NOAA Chemical Sciences Laboratory chemiluminescence instrument, and NO2 was measured with the Berkeley Thermal Dissociation Laser-Induced Fluorescence instrument. On the GV aircraft, CO was measured by the NCAR Aero Laser Vacuum Ultraviolet fluorescence instrument, and O3, NO, and NO2 were observed by the NCAR chemiluminescence instrument. NO2 is converted to NO by UV photolysis
prior to the chemiluminescent measurement. See Barth et al. (2015) for references describing these instruments. Huntrieser et al. (2016a) describe the characteristics of the 29–30 May storm and discuss the Falcon O₃ observations.

3.2. 29–30 May 2012 Oklahoma Severe Storm

On the morning of 29 May 2012 conditions were very unstable and supportive for convective development within the Oklahoma domain. The 12:00 UTC sounding from Norman, Oklahoma, indicated the wind profile generally had a westerly component with veering from the surface to 600 hPa. A dryline was situated across the Texas panhandle, and a frontal system stretched southwestward from the Ohio River Valley with the southern portion moving northward as a warm front over Oklahoma and Kansas. By 21:10 UTC the front became quasi-stationary and two isolated cells developed to its south along the Kansas-Oklahoma border and ahead of the dryline. Over the next several hours both storms formed mid-level mesocyclones (DiGangi et al., 2016) and evolved into a line of supercells. The supercell line continued in an east-southeast direction, and new, non-supercellular storm cells formed both behind the supercell line and far downstream within the merged forward anvil region of the supercells (DiGangi et al., 2021). The combined anvil-initiated storm line and remnant supercells subsequently coevolved to form a MCS that moved through central Oklahoma during the period 03:00–04:00 UTC on 30 May.

3.3. Lightning and NOₓ Observations in the 29–30 May Storm

Cummings et al. (2024) analyzed the LMA data in terms of the geographic distribution of flashes and in terms of the time series of flash rates for the 29–30 May storm. Flashes were required to have at least 10 very high frequency (VHF) sources detected by 6 or more stations, and a reduced chi-squared solution error of less than 5.0. Figure 1 in Cummings et al. (2024) shows the progression over time of lightning initiation points from storm initiation just south of the Oklahoma-Kansas border at 21:10 UTC 29 May until dissipation around 0420 UTC 30 May. The convection developed between the northern edges of the LMA network's 2D (200 km radius of network center) and 3D (100 km radius from network center) lightning detection coverage. As the storm system moved east-southeastward, it remained within the coverage of the network for almost its entire life and was within the 3D coverage area between 23:40 and 03:00 UTC. The total flashes observed over the lifetime of the convection numbered 31,633 with an average total flash rate of 75.5 flashes min⁻¹. The total flashes numbered 15,060 during the 5-hr period of our storm simulation (20:00–01:00 UTC observed time), with a mean total flash rate of 68.5 total flashes min⁻¹. This period encompasses the aircraft sampling of the storm. The 1-min data were binned into 10-min intervals and plotted (Figure 1). The observed total flash rate time series shows several peaks as the flash rate increased near zero at 21:30 UTC to the maximum (> 2,600 flashes 10-min⁻¹) at 01:40 UTC (not shown), shortly after the aircraft departed from the storm. These peaks of roughly 100, 300, 800, and 2,200 total flashes 10-min⁻¹ occurred at 22:00, 22:50, 23:50, and 01:00 UTC. Following the primary peak, the observed flash rate decreased at a faster rate than it increased (see Cummings et al., 2024). The 29–30 May 2012 convection contained high flash rate supercells with small flash extents (Barth et al., 2015). DiGangi et al. (2016) also showed that the Kingfisher storm, which was within the southern portion of the convection analyzed in this case study, had an abundance of small flashes, especially during its mature phase, when flash rates were > 100 flashes min⁻¹. The 10-min time-evolving (IC:CG) flash ratios were calculated over the life of the storm using the observed NLDN CG and LMA total lightning flash data (Figure 1). The mean time varying IC:CG ratio over the storm lifetime is 2.73 ± 2.51, while the mean for the 5-hr simulation period is higher (3.93 ± 2.22). Cummings et al. (2024) also analyzed the LMA VHF sources in terms of their altitude and associated radar reflectivity (see Cummings et al., 2024, Figure 9). The greatest percentage of sources were located between 8 and 10 km and with reflectivities between 25 and 35 dBZ. The LMA source data were also used in the calculation of flash extent, vertical profiles of which were used in determining the vertical placement of LNOₓ in the model. Fuchs et al. (2015) provide further details on the use of LMA sources in flash channel construction and errors involved in these calculations.

During this storm the DC-8 focused first on low-level storm inflow and then spiraled up to begin sampling outflow, while the GV concentrated on high-altitude outflow. Figure 2 shows the time series of NOₓ observed by the GV and DC-8 during the flight shown as 10-s and 1-min averages. Figure 3 maps the 1-min average NOₓ data along the transects of the anvil conducted by the DC-8 and GV aircraft. The GV sampled storm outflow at seven altitudes between 10 and 12.5 km. The initial transects were downstream of the anvil while the storm was moving closer. The largest in-anvil peak NOₓ reached 2.8 ppbv at 12 km near the northern edge of the anvil.
The DC-8 achieved two anvil transects with 10-s NO$_x$ reaching 5.7 ppbv at 11 km near new cells that developed within the anvil. NO$_x$ measurements were not available for the Falcon for this research flight.

4. Model Setup

4.1. Cloud-Resolved Simulation of 29–30 May Oklahoma Storm and Chemistry

The WRF-Chem (Version 3.6.1; NCAR, 2014) model was used to simulate the 29–30 May Oklahoma severe storm and associated chemistry. The model domain contains 480 x 420 x 89 grid points in the x, y, and z directions with 1-km horizontal resolution and variable vertical resolution from 50 m near the surface to 250 m at the top of the domain (50 hPa). The southwest corner of the domain is located at 33.8196°N, 99.7726°W and the northeast corner is located at 38.1354°N, 95.0998°W. An initial 8-hr simulation ran from 18:00 UTC on 29 May to 02:00 UTC on 30 May using 480 processors on the NCAR Yellowstone supercomputer. The simulation does not extend past 02:00 UTC because we are interested in the time period when lightning would be influencing the NO$_x$ mixing ratios sampled by the DC3 aircraft in the anvil outflow. For subsequent simulations used to constrain the LNO$_x$ production per flash, a restart file was used at 21:00 UTC to reduce computation time since the observed convection began just after 21:00 UTC. The simulation is integrated with a 3 s time step, while the lightning parameterization is called every 6 s.
The initial model configuration and physics and chemistry options were based on Bela et al. (2016). Meteorological initial and boundary conditions came from the 6-hr interval North American Mesoscale Analysis (NAM-ANL; NOAA, 2012) at 12 km resolution. The constant initial and boundary chemistry conditions for all the WRF-Chem simulations are reset at 21 UTC using a combination of data from the DC-8 profile (i.e., for the boundary layer and free troposphere) and the Model for Ozone and Related chemical Tracers version 4 (MOZART). Table 1 provides the final model physics and chemistry options selected for this analysis. Detailed descriptions of some of the selected schemes are provided below.

We selected the MOZART chemistry mechanism to be consistent with the incoming initial and boundary conditions. The MOZART chemical scheme is connected to WRF-Chem via the Kinetic Pre-Processor (KPP) and was run combined with the Georgia Tech/Goddard Chemistry Aerosol Radiation and Transport (GOCART) aerosol scheme (MOZCART). GOCART is coupled to the radiation schemes in WRF-Chem, such that the direct effect of aerosols can be considered. However, it is not connected with the cloud microphysics schemes in the WRF-Chem version used for this case study.

Cummings et al. (2024) used two methods (convective damping and Lightning data assimilation (LDA)) to guide the model when simulations did not reproduce the observed convection in the correct location or time. Details of these two schemes are provided by Cummings et al. (2024). We implemented these techniques in this cloud-resolved simulation to try to bring the simulated storm closer to observations. Using the damping and LDA schemes. Cummings et al. (2024, Figures 4 and 5) demonstrated that the model depiction of radar reflectivity over the lifetime of the storm from initiation through dissipation compared well with observations. Figure 4 of this paper shows the model depiction of total condensate at 10.5 km (anvil level) at two times (soon after initiation and at the mature stage) alongside visible satellite imagery. The model estimates of anvil size are similar to that seen on the satellite images.

Figure 3. NOx observations from GV (X) and DC-8 (+) aircraft transects across the anvil of the 29–30 May 2012 severe storm superimposed on observed NEXRAD radar reflectivity. Upper left, 22:10 UTC; upper right, 23:00 UTC; lower left, 23:50 UTC; lower right, 00:30 UTC.
### Table 1

<table>
<thead>
<tr>
<th>WRF model options</th>
<th>Name of scheme</th>
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<tr>
<td>Microphysics</td>
<td>Morrison 2-moment scheme</td>
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<td>Planetary boundary layer</td>
<td>Yonsei University (YSU) scheme</td>
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<tr>
<td>Surface layer</td>
<td>MM5 similarity scheme</td>
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<tr>
<td>Land surface</td>
<td>Unified Noah land-surface model</td>
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<tr>
<td>Radiation (shortwave and longwave)</td>
<td>Rapid radiative transfer model for GCMs (RRTMG) scheme</td>
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<tr>
<td>Cumulus parameterization</td>
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</tr>
<tr>
<td>Convective damping</td>
<td>21:00–21:20 UTC (Li et al., 2017)</td>
</tr>
<tr>
<td>Lightning data assimilation (LDA)</td>
<td>21:30–23:00 UTC (Fierro et al., 2012, 2014, 2015)</td>
</tr>
<tr>
<td>Lightning flash rate parameterization</td>
<td>• ICEFLUX scheme (Finney et al., 2014)</td>
</tr>
<tr>
<td>LNO$_2$ vertical and horizontal distributions</td>
<td>• IC:CG ratios based on LMA and NLDN observations</td>
</tr>
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<td>Temperature of peak of LNO$_2$ vertical distribution for lightning</td>
<td>• Radar reflectivity ≥20 dBZ</td>
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<tr>
<td>Moles NO emitted per flash</td>
<td>• IC peak 45°C</td>
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<tr>
<td>Chemical mechanism</td>
<td>• CG peak 40°C</td>
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<td>Wet scavenging and ice retention</td>
<td>• IC 82; CG 82</td>
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<td></td>
<td>MOZART (MOZART chemistry and GOCART aerosols)</td>
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<td></td>
<td>Assume zero retention for trace gases</td>
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#### 4.1.1. Lightning Flash Rate Parameterization in WRF-Chem

Cummings et al. (2024) found that vertical cross sections of WRF-Chem model frozen hydrometeors (graupel, snow, ice) did not match the altitude ranges of proxy hydrometeor observations derived by DiGangi et al. (2016). Scaling factors as a function of altitude for the hydrometeors were developed to adjust the WRF-Chem output to better agree with the proxy observations. The scaled hydrometeors were used only in the FRPS calculations.

Cummings et al. (2024) determined that the ICEFLUX FRPS of Finney et al. (2014) yielded the best comparison with the LMA flash rate data for the 29–30 May storm for the period from storm initiation through the end time of the aircraft sampling (0100 UT) in terms of total predicted flashes, centered root mean squared error per 10-min, and mean bias per 10-min. The ICEFLUX scheme is based on the upward flux of cloud ice at 440 hPa. However, Cummings et al. (2024) found that for this storm evaluating the flux at 390 hPa produced a better comparison with observed flash rates. Figure 1 shows the time series of flash rates produced by the modified ICEFLUX scheme over this time period. The parameterized flash rates increase more rapidly than observed at the beginning of the storm, but after 00:00 UTC 30 May they become less than observed. Over the time period from storm initiation to 01:00 UT, the ICEFLUX scheme produced 14,419 flashes, which is within 4.5% of the observed 15,060 flashes.

The WRF-Chem model provides several options for controlling the IC:CG ratio. One method uses the coarsely prescribed ratios based on the 1995–1999 NLDN/Optical Transient Detector (OTD) climatology from Boccioppio et al. (2001). For this case study, this method would assign an IC:CG ratio of 3.55 to the central United States (90–105°W). We replaced this single climatological value with the actual observed 10-min time evolving IC:CG ratios (Figure 1) only within the area of the associated 10-min moving spatial mask that surrounds the storm system of interest. These values ranged from near zero to 8, and averaged 3.93 over the period ending at 01:00 UTC, which is slightly larger than the climatological value.

#### 4.1.2. Lightning NO$_2$ Parameterization in WRF-Chem

By default, WRF-Chem uses Gaussian vertical distributions of lightning channel segments for CG and IC flashes (DeCaria et al., 2000) for thunderstorm simulations at cloud-resolved scales. In this study, these default distributions were replaced with the observed vertical profiles constructed using the flash extent data from the central Oklahoma LMA network during the 29–30 May convective event. The observed CG and IC vertical distributions at 1-min resolution showed that the profile shape and the altitude of the peak in flash channel lengths varied over
the lifetime of the convection; however, the mean profiles for both flash types were unimodal nearly Gaussian distributions (Figure 5). The CG vertical profile maximizes at a considerably colder temperature and higher altitude (−40°C or 9–10 km) than the default vertical distribution (15°C or 6 km). The observed and default IC vertical profiles both peak at 45°C (10–11 km), but the observed profile does not have a secondary lower-altitude peak as in the default distribution. The observed vertical profiles of flash extent for the 29–30 May storm system also peaked at a higher altitude than is typical for Oklahoma storms. The composite flash extent vertical profile for warm season 2011 storms in Oklahoma is illustrated by Fuchs and Rutledge (2018), indicating a peak between 6 and 9 km with this altitude range containing slightly over 50% of the flash extent. However, the profile peak altitudes for the 29–30 May storm are more consistent with those derived from North Alabama LMA data by Mecikalski and Carey (2017) for Alabama supercell storms.

Figure 4. Comparison of the anvil area from the 29–30 May 2012 severe supercell system detected by GOES-13 (NCAR, 2012a, 2012b) at (a) 21:30 UTC and (c) 23:40 UTC observed time and by the WRF-Chem model-simulated total hydrometeors (g kg⁻¹) at 10.5 km altitude at (b) 22:30 UTC and (d) 00:40 UTC model time. The dashed black box in each image highlights the portion of the convection that is the focus of the observed and modeled storm at the selected time.
The vertical distribution of LNO$_2$ production involves the use of the observed flash extent profiles convolved with atmospheric pressure. The horizontal placement of the LNO$_2$ follows the bulk approach and is injected within the 20 dBZ reflectivity contour (DeCaria et al., 2005; Ott et al., 2010). The radar reflectivity criterion is not modified from the default value because the vast majority (all but 2%) of the observed LMA sources are within the 20 dBZ contour in this storm (see Figure 9 of Cummings et al., 2024). This high percentage within the 20 dBZ contour may be typical for supercells. Mecikalski and Carey (2017, 2018) showed that supercells in North Alabama had a higher percentage (80%–88%) of flashes at locations with radar reflectivities $>20$ dBZ than other storm types.

4.2. Regional Model Simulation of Downwind Storm Outflow Chemistry

We conducted a WRF-Chem simulation at 36 km horizontal resolution to examine the upper tropospheric downwind transport and chemistry from the 29–30 May supercell storm. The model setup details are listed in Table 2. Convection was parameterized using the Grell-Freitas (GF) scheme with Kain-Fritsch (KF) closure, as this combination yielded the best results for predicted precipitation for this storm (Li et al., 2018). Other physical parameterizations used included the Mellor-Yamada-Janjic scheme (Janjic, 1994) for the boundary layer, two-moment Morrison microphysics scheme (Morrison et al., 2009), the Rapid radiative transfer model for GCMs scheme (Iacono et al., 2008) for longwave and shortwave radiation, and the Noah scheme (Koren et al., 1999) for land surface processes.

The chemistry option was MOZART chemistry using the KPP library. Photolysis rates were calculated using the F-TUV photolysis scheme (Tie et al., 2003). Fire emissions were calculated from the FINN data. The 2011 NEI data were used to create anthropogenic emissions. Anderson et al. (2014) showed that NEI NO$_x$ emissions were overestimated by 51%–70%, and Travis et al. (2016) found that NEI overestimated the NO$_x$ emission by 30%–60%. Therefore, we reduced the NEI NO$_x$ emission by the mean of these four values (50%) in our simulation. We used the MEGAN v2.04 to generate biogenic emissions. Aircraft emission data were obtained from Baughcum et al. (1999). LNO$_2$ production was set for both cloud-to-ground and intracloud flashes at the number of moles per flash determined from the cloud-resolved modeling (see Section 5.1.2). The vertical distributions of the LNO$_2$ emissions were the same as used in the cloud-resolved modeling (Figure 5). Flash rates were parameterized using the PR92 (Price & Rind, 1992; Wong et al., 2013) lightning option which is the recommended method for predicting lightning flash rate for parameterized convection in WRF-Chem. This scheme is based on the level of neutral buoyancy from the convective parameterization. However, this scheme severely underpredicted the flash rates for this very high flash rate storm. Therefore, we used a flash rate adjustment factor of 17 (ratio of observed flashes to unadjusted flashes predicted by the model) to get the best simulation of the Earth Networks Total Lightning Network (ENTLN) observed storm total flashes. Figure 6 shows the time series of ENTLN flashes and the adjusted model flash rates for a larger region and longer time period than used in the cloud-resolved analysis.

We used MOZART-4 (Emmons et al., 2010) to create the chemical initial and boundary conditions. In order to have a better simulation of inflow, we adjusted the initial conditions to better match aircraft observations using the equations listed in Table 3. The resulting model-simulated inflow trace gas mixing ratios also are compared with aircraft data in the same table. The differences between the simulation and observation of all the species in low-level inflow were within 10% of the aircraft measurements, except H$_2$O$_2$. However, the overestimate of H$_2$O$_2$ in the inflow will not affect the H$_2$O$_2$ in the outflow due to its high solubility and short lifetime in the lower troposphere. NO$_x$ in the inflow was within 10% of observations without adjustment. Li et al. (2019) tested ice retention factors ($r$) for use with parameterized convection. Here we use the best estimate of these factors, which are $r = 0$ for CH$_2$O and CH$_2$OOH, $r = 0.1$ for H$_2$O$_2$, $r = 1$ for HNO$_3$, and $r = 0.02$ for SO$_2$, as well as an improved
conversion rate for cloud water to precipitation at below freezing temperatures in our simulation to study the ozone change downwind of deep convection.

The main feature of the downwind chemistry in which we are interested is the net $O_3$ production rate over the approximately 24 hr of transport that occurred between the aircraft sampling of anvil outflow over Oklahoma and the sampling the following day over the southern Appalachians. Considering this, we calculated the 24-hr net ozone production between 00:00 UTC 30 May (model time: 01:20 UTC 30 May) and 00:00 UTC 31 May (model time: 01:20 UTC 31 May) at all grid points in the downwind region of the flights. This calculation was performed

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**Table 2**

**WRF-Chem Model Physics and Chemistry Options Selected for Cloud-Parameterized Simulation of the 29–30 May 2012 Oklahoma Storm**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Meteorology initial/boundary conditions</td>
<td>NAM 18 UTC</td>
</tr>
<tr>
<td>Chemistry initial/boundary conditions</td>
<td>MOZART scaled</td>
</tr>
<tr>
<td>Grid resolution</td>
<td>36 km</td>
</tr>
<tr>
<td>Vertical levels</td>
<td>90</td>
</tr>
<tr>
<td>Time step</td>
<td>120 s</td>
</tr>
<tr>
<td>Cumulus parameterization</td>
<td>GF with KF closure</td>
</tr>
<tr>
<td>Microphysics</td>
<td>Morrison</td>
</tr>
<tr>
<td>PBL</td>
<td>MYJ</td>
</tr>
<tr>
<td>Longwave radiation</td>
<td>RRTMG</td>
</tr>
<tr>
<td>Shortwave radiation</td>
<td>RRTMG</td>
</tr>
<tr>
<td>Lightning schemes</td>
<td>Price and Rind (1992; PR92) based on level of neutral buoyancy (Wong et al., 2013)</td>
</tr>
<tr>
<td>Cloud top height adjustment</td>
<td>0</td>
</tr>
<tr>
<td>Flashrate factor</td>
<td>17</td>
</tr>
<tr>
<td>LNO$_2$ scheme</td>
<td>Combined IC and CG flashes single-mode vertical distributions with LMA flashes vertical profile</td>
</tr>
<tr>
<td>Moles of NO emitted per IC flash</td>
<td>82</td>
</tr>
<tr>
<td>Moles of NO emitted per CG flash</td>
<td>82</td>
</tr>
<tr>
<td>Fire emissions</td>
<td>FINN</td>
</tr>
<tr>
<td>Anthropogenic emissions</td>
<td>NEI with NO$_x$ reduced by 50%</td>
</tr>
<tr>
<td>Biogenic emissions</td>
<td>MEGAN v2.04</td>
</tr>
<tr>
<td>Chemistry option</td>
<td>MOZART</td>
</tr>
</tbody>
</table>

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**Figure 6.** Vertical axis shows Earth Networks Total Lighting Network measured flashes (blue) and model simulated flashes (orange) versus time (UTC; horizontal axis). Data are for 10-min intervals covering the region 35° to 38.5°N and 95.5° to 100.5°W (a larger region and longer time period than considered in the cloud-resolved analysis).
Table 3
Initial Condition Formulation Equations With Inflow Observations and Resulting Inflow Mixing Ratios in the WRF-Chem Simulation

<table>
<thead>
<tr>
<th>Species</th>
<th>Initial condition equations</th>
<th>Observation (ppbv)</th>
<th>WRF-chem inflow (ppbv)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO</td>
<td>( C_C^{x(72)} ) ( 2H/100 )</td>
<td>136.8 1.1</td>
<td>141.4 1.4</td>
</tr>
<tr>
<td>CH(_2)O</td>
<td>( C_C^{y(14)} H ) ( * 0.8 ) ( 1 )</td>
<td>5.2 0.3</td>
<td>4.9 0.2</td>
</tr>
<tr>
<td>CH(_2)OOH</td>
<td>( C_C^{y(14)} H ) ( * 0.25 ) ( 1 )</td>
<td>1.6 0.1</td>
<td>1.6 0.0</td>
</tr>
<tr>
<td>HNO(_3)</td>
<td>( C_C^{y(58)} 3H/100 )</td>
<td>0.6 0.1</td>
<td>0.6 0.0</td>
</tr>
<tr>
<td>O(_3)</td>
<td>( C_C^{y(60)} 10H/100 )</td>
<td>54.9 1.9</td>
<td>59.5 0.8</td>
</tr>
<tr>
<td>H(_2)O(_2)</td>
<td>0.3( C_C )</td>
<td>2.5 0.2</td>
<td>6.6 0.0</td>
</tr>
</tbody>
</table>

Note. \( C_C \) is the inflow IC from MOZART-4 output; \( C \) is the adjusted inflow IC; \( H \) is altitude in km. Equations applied from surface to 14 km except for \( O_3 \), which was applied from surface to 4 km.

by creating an \( O_3 \) production tracer to which was added the chemical tendency for \( O_3 \) from the model at each time step beginning at 00:00 UTC (model time: 01:20 UTC). The tracer accumulation was performed for each grid cell over the period of downwind transport. The net \( O_3 \) production was evaluated using the values of this tracer over the model domain at the end of the 24-hr simulation.

5. Results

5.1. Cloud-Resolved Chemistry

To estimate the mean LNO\(_3\) production per flash for the observed 29–30 May DC3 storm using WRF-Chem, results from several cloud-resolved simulations are evaluated. In this sub-section we first describe the eight simulations performed. We then show that the convective transport of CO and \( O_3 \) for those eight simulations give consistent results before investigating which of the LNO\(_3\) simulations performed best compared to anvil \( N_2O_5 \) observations.

The simulations (Table 4) included one run with no lightning chemistry and seven runs with different LNO\(_3\) production scenarios. All seven simulations contained the FRPS based on ICEFLUX. Prior WRF-Chem cloud-

Table 4
The Mean Trace Gas Mixing Ratios Over the Four Model Layers the Aircraft Flew Within (10.72–10.97, 10.97–11.21, 11.71–11.95, and 11.95–12.20 km)

<table>
<thead>
<tr>
<th>LNO(_3) production scenario</th>
<th>Aircraft observed mean mixing ratios (ppbv)</th>
<th>Model-simulated mean mixing ratios (ppbv)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>CO</td>
<td>( O_3 )</td>
</tr>
<tr>
<td>IC 0</td>
<td>118.3</td>
<td>5.5</td>
</tr>
<tr>
<td>IC 125; DeCaria vertical; Boccippio IC:CG</td>
<td>118.5</td>
<td>4.9</td>
</tr>
<tr>
<td>IC 125</td>
<td>118.4</td>
<td>5.1</td>
</tr>
<tr>
<td>IC 500</td>
<td>117.9</td>
<td>4.6</td>
</tr>
<tr>
<td>IC 107</td>
<td>118.1</td>
<td>5.0</td>
</tr>
<tr>
<td>IC 82</td>
<td>117.6</td>
<td>5.0</td>
</tr>
<tr>
<td>IC 38; CG 604</td>
<td>118.1</td>
<td>5.4</td>
</tr>
<tr>
<td>IC 21; CG 328</td>
<td>118.5</td>
<td>4.7</td>
</tr>
</tbody>
</table>

Note. \( CO \) and \( O_3 \) model values taken over (10.48–10.72, 10.72–10.97, 11.46–11.71, and 11.71–11.95 km). The asterisk indicates the observed LNO\(_3\) is an estimation based on mean aircraft observed total \( N_2O_5 \) and the mean model-simulated in-cloud background \( N_2O_5 \). Unless noted otherwise, each LNO\(_3\) production scenario used the observed flash channel vertical distribution and IC:CG ratio.
resolved simulations (Bela et al., 2016) of the 29–30 May case study used the default model parameters for the fractional CG and IC flash channel vertical profiles (DeCaria et al., 2000) and the climatological IC:CG ratio (Boccioppio et al., 2001) with the maximum vertical velocity ($w_{\text{max}}$) FRPS with a 0.106 adjustment factor. This analysis tentatively suggested an LNO$_3$ production scenario of 125 mol flash$^{-1}$. However, since observations of the vertical distributions of CG and IC flash channels and of the IC:CG ratio are available for the 29–30 May storm, subsequent simulations used this information.

The subsequent simulations (Table 4) were used to determine a final LNO$_3$ production scenario based on the best fit of the anvil NO$_3$ model output with the DC3 aircraft observations. We first tested the 125 mol flash$^{-1}$ scenario with the observed IC:CG ratios and vertical distributions of flash channels. The previously suggested 500 mol per CG and IC flash (Ott et al., 2010) scenario was also tested. Pollack et al. (2016) concluded 107 mol flash$^{-1}$ was appropriate for the 29–30 May case using a volume-based method to analyze DC3 aircraft measurements, and this scenario was tested in the model. A comparison of the model with 107 mol flash$^{-1}$ and aircraft NO$_3$ mixing ratios suggested the LNO$_3$ production scenario should be lower than that value. Additional simulations with smaller production values were tested, and a scenario with 82 mol flash$^{-1}$ achieved the best match with observations. The Koshak et al. (2014) scenario of 604 mol per CG flash and 38 mol per IC flash, which yields a mean 151 mol flash$^{-1}$ using the 29–30 May observed mean IC:CG ratio of about four was also used. When the mean Koshak et al. (2014) value of 151 mol flash$^{-1}$ is adjusted to 82 mol flash$^{-1}$, but maintaining the same ratio of NO production per CG flash and per IC flash, this suggested a scenario of CG 328 mol flash$^{-1}$ and IC 21 mol flash$^{-1}$ to test. The detailed results of these six tests are presented in Section 5.1.2, but, first, Section 5.1.1 discusses the convective transport of CO and O$_3$ in the 29–30 May storm under the various LNO$_3$ scenarios.

5.1.1. Convective Transport of CO and O$_3$

To confirm convective transport is consistent among the eight simulations, measurements and model results for CO and O$_3$ in the anvil outflow are compared. The anvil outflow most directly from the storm cores was sampled by the DC-8 between 10.72 and 11.21 km from 23:48:30 to 23:58:30 UTC and by the GV between 11.71 and 12.20 km from 23:59:30 to 00:23:30 UTC (see Figures 2 and 3). Each altitude interval corresponds to two model layers. These GV and DC-8 anvil outflow sampling periods were defined by Fried et al. (2016) as periods when aircraft wind vectors were pointed away from storm cores toward the aircraft positions when flying through the anvil cloud. The periods were divided into 10-min intervals to correspond with the 10-min model output. Model data points were required to be within anvil cloud outflow (total hydrometeors >0.01 g kg$^{-1}$) and within tropospheric air (O$_3$:CO ratio <1.25; Hudman et al., 2007) to be included. Since the observed and simulated anvil clouds were of similar area, outflow sampling boxes of comparable size were identified in the model-simulated convection. The box locations were adjusted in the horizontal plane to represent a similar distance from the center of the observed high composite reflectivity, as well as a region with similar features as the observed convection. The only exception was the model outflow box from 23:50 to 00:00 UTC relative to the DC-8 observations. The southwest portion of the flight track was located between 10.97 and 11.21 km and the northern half was between 10.72 and 10.97 km. In this lower layer the model-simulated wind speed was slightly weaker, and the LNO$_3$ plume was thicker than observed; therefore, this layer was shifted to a region farther east in the model than the observations. Increasing the distance of the lower layer from the model-simulated convective core during this time interval placed the northern portion of the outflow box in a region of the model anvil that is more representative of the observed anvil environment.

The 1-s DC-8 and GV aircraft data are averaged over 5 and 4 s, respectively, to yield observations of similar horizontal scale for comparison with the 1 km 1 km horizontal resolution of the model data. A direct comparison of the aircraft observed and model-simulated CO and O$_3$ mixing ratios within the same layers indicate the convective transport from the boundary layer is underestimated in the model (Figures 7a and 7b). CO is overestimated by the model at DC-8 altitudes and underestimated at GV altitudes. The opposite is true for O$_3$. Too much boundary layer air was deposited by the model at DC-8 levels and insufficient boundary layer air was delivered to GV levels. Mixing ratios from one model layer (i.e., 0.25 km) beneath each layer at which the aircraft flew (i.e., 10.48–10.72 and 10.72–10.97 km for the DC-8; 11.46–11.71, and 11.71–11.95 km for the GV) provides a much better match to the observations (Figures 7c and 7d). Using these layers as a correction for the underestimated vertical transport, the overall mean model-simulated mixing ratios from these four model layers are within 1% and 3% of the aircraft observed CO (118.7 ppbv) and O$_3$ (86.1 ppbv), respectively (Table 4). This is true for each of the WRF-Chem simulations run with and without lightning chemistry.
Horizontal and vertical cross-sections of the model-simulated CO and O₃ from the 82 mol NO per flash scenario are shown in Figures 8 and 9 for model times 23:50 UTC and 01:40 UTC, respectively. These figures illustrate some of the features of convective transport associated with this storm. The convective simulation suggests that lower CO and higher O₃ mixing ratios are transported downward around the south and west sides of the anvil. For this DC3 case, Huntrieser et al. (2016a) illustrated measurements of O₃-rich stratospheric air on the edge of the anvil to the south and west (100–180 ppbv) of the storms as well as other locations to the east (100–220 ppbv) of the storm. Li et al. (2017) simulated the vertical flux divergence in three DC3 convective events using WRF-Chem at different spatial scales, including our 29–30 May case study, and captured a similar O₃ enhancement 2–3 km below the tropopause. During DC3, research aircraft often encountered O₃-rich stratospheric air wrapped around the cloud edge of severe storms (Huntrieser et al., 2016a, 2016b; Pan et al., 2014).

Vertical cross-sections show model-simulated boundary layer CO (>130 ppbv) and O₃ (<65 ppbv) are present in the convective updrafts (Figures 8 and 9). A layer of higher CO mixing ratios (>120 ppbv) also exists at 10.5 km, as well as within the anvils. Minima in O₃ mixing ratios are visible in the anvil for several hours in the 5-hr simulation. In the lower portion of the anvil (8–10 km) O₃ decreased by 20 ppbv due to convective transport. Above 10.8 km within the convective system, where the initial mixing ratios exceeded 100 ppbv, the mean O₃ mixing ratio decreased by 40 ppbv by the end of the simulation at 02:00 UT model time. A comparison of the decrease in O₃ with and without lightning chemistry indicates that titration by LNO₃ only accounts for roughly a 1–2 ppbv decrease at most within the anvil. This suggests the upward transport of lower O₃ mixing ratio boundary layer air into the cloud is responsible for almost all of the decrease in O₃ at anvil levels.

5.1.2. Lightning NO₃ Simulations

Since LNO₃ is primarily injected into the middle to upper troposphere, it is not influenced by convective transport to the same degree as CO and O₃. For the 29–30 May event, the peaks in flash channel lengths (both CG and IC) and LNO₃ occur at 9.5 and 10.5 km, respectively. Since LNO₃ is inserted into the model following a bulk

**Figure 7.** Aircraft observed and model-simulated mean (a) CO and (b) O₃ mixing ratios within the same layers. Model-simulated mean (c) CO and (d) O₃ mixing ratios moved up 0.25 km. The standard deviation is represented by solid black lines for the observations and dotted color lines for the model.
approach as opposed to a filament approach, it is not realistic to expect the model to capture the proper mixing ratios at specific locations. Therefore, the NO₃ and LNOₓ mixing ratios were only compared with observations in a statistical sense using means and standard deviations within the model layers where the aircraft flew (Figure 10). The distribution and variability of NO₃ and LNOₓ in the horizontal and vertical are demonstrated in the cross sections in Figures 8 and 9.

With lightning chemistry turned off, the overall mean NOₓ from the four model layers (0.6 ppbv) underestimates the aircraft observed NOₓ (1.4 ppbv) by 0.8 ppbv (Table 4). The scenario with 125 mol LNOₓ per flash that incorporates the default model parameters for the fractional CG and IC flash channel vertical profiles (DeCaria et al., 2000) and IC:CG ratio (Boccippio et al., 2001) underestimates the aircraft observed NOₓ by 15%. However, the default profiles placed LNOₓ lower in the troposphere than observed based on the lightning observations. Specifically, the default CG channel lengths peak 3.5 km below the observed peak for this storm system, while the default IC channel length has a secondary peak that maximizes at 6 km that is not present in the observations (single 10.5 km peak). When the 125 mol flash⁻¹ scenario is run with the observed flash channel profiles and IC:CG ratios, the aircraft observations are exceeded by 35% indicating that the PE of LNOₓ is likely less than 125 mol flash⁻¹. The best agreement with aircraft observations in the anvil outflow is found for a value of 82 mol per CG and IC flash, which is near the low range of estimates obtained by Pollack et al. (2016) using a volume-based estimate based on anvil observations. They obtained a mean PE of 107 mol flash⁻¹, which is 30% more than the PE found in this study. Using 107 mol flash⁻¹ in this study led to an overestimate of NOₓ by 20%. Based on the overall mean from the four model layers, the model-simulated NOₓ mixing ratio (1.3 ppbv) for the 82 mol flash⁻¹ scenario is within 7% of the aircraft observational mean (1.4 ppbv). This LNOₓ production
Figure 9. Similar to Figure 8, except the model-simulated trace gases represent 01:40 UTC model time.

The two scenarios in which the LNO₂ production per CG flash was substantially greater than that for an IC flash both overestimated the observed NO₃. The scenario using the IC and CG values (CG: 604; IC: 38 mol fl⁻¹) from Koshak et al. (2014) overestimated by 86%, which is likely due to the mean (151 mol fl⁻¹) being too large. The scenario that was adjusted to use a mean of 82 mol fl⁻¹ (CG: 328; IC: 21 mol fl⁻¹) was within 10% of the aircraft observed NO₃, averaged over all four layers; however, the largest overestimate is within the 10.72–10.97 km layer (80%), while biases in the mean model-simulated NO₃ within the other three model layers ranged from 20% to 11%. This results in a poorer simulation than the scenario that uses 82 mol per CG and IC flash in which each of the four layers are within 20% of the observations and the model NO₃ averaged over these layers underestimates the observations by only 7%. This suggests that mean LNO₂ production by individual CG and IC flashes is similar in this storm. It is unclear if storm-to-storm variability, regional differences, or another factor are the cause of the large differences in the CG:IC PE ratio between this study and Koshak et al. (2014).

All further analyses and discussions will be based on the LNO₂ production scenario of 82 mol NO per CG and IC flash, unless otherwise noted. In the model the NO production from lightning is added to the NO species variable that is chemically active, but LNO₂ is also tracked separately as a passive species. For the 82 mol NO per flash scenario the resulting passive LNO₂ is 1.2 ppbv. Since the aircraft cannot directly measure LNO₂ in the anvil outflow, the “observed LNO₂” is estimated by taking the difference between the total NO₂ observed by the aircraft (1.4 ppbv) and the model-simulated in-cloud NO₂ from the no-lightning simulation (0.6 ppbv) resulting from convective transport from the boundary layer. Therefore, our estimate of mean “observed LNO₂” in the cloud is at a minimum 1.4 minus 0.6 or 0.8 ppbv. This value, however, is affected by chemical loss of NO₂ during the course
The solid line represents the observed mixing ratio of NO on 3 May and the dotted line represents the observed mixing ratio of NO on 1 May. The figures show that the model-simulated LNO\textsubscript{3} mixing ratio is higher than the observed mixing ratio during the storm event. The model results suggest that the LNO\textsubscript{3} production is significantly enhanced during the storm event. The enhancement is likely due to the atmospheric conditions during the storm event, which can lead to increased nitrate formation.

In the model simulation, NO\textsubscript{3} and LNO\textsubscript{3} maxima were co-located with the O\textsubscript{3} minimum in the upper portion of the convective core (> 7 km), as well as the anvil. Here the mixing ratios for NO\textsubscript{3} and LNO\textsubscript{3} exceed 2 ppbv. While vertical cross-sections suggest a small amount of boundary layer NO\textsubscript{3} is transported into the upper region of the convective cells, the model-simulated LNO\textsubscript{3} appears to make up 88% of the NO\textsubscript{3} within the anvil.

The LNO\textsubscript{3} PE values obtained in this study were obtained by tuning the LNO\textsubscript{3} source to ensure that model values match observations. Thus, incomplete model chemistry and/or measurement interferences add uncertainty to the results. Nault et al. (2016, 2017) found that the lifetime of NO\textsubscript{3} in the near field of deep convection is much shorter than had been assumed. They indicate that the lifetime may be 3 hr in the near-field outflow from thunderstorms due to rapid production of methyl peroxy nitrate (MPN) and alkyl nitrates, but the lifetime becomes much longer as the air moves downwind. The MOZART-4 mechanism does not contain MPN, and production of alkyl nitrates is not well represented. These findings suggest that our model-based estimate of LNO\textsubscript{3} PE may be biased low because of a lack of adequate accounting for these nitrate compounds, leading to a need for a resulting larger value of LNO\textsubscript{3} production per flash. Nault et al. (2017) indicate that models without the revised chemistry mentioned above may underestimate LNO\textsubscript{3} PE by up to 33%. Brune et al. (2021) found large enhancements of OH and HO\textsubscript{2} in DC3 storms that were linked with visible and subvisible lightning flashes. The lightning-enhanced HO\textsubscript{2} (LNO\textsubscript{3}) leads to increased HONO production (a few pptv in the presence of the 1–2 ppbv LNO\textsubscript{3} observed in this case). Again, our model LNO\textsubscript{3} production calculation could have a small low bias due to unaccounted HONO production. Shah et al. (2023) concluded that there was interference in the DC3 TD-LIF measurements of NO\textsubscript{2}, presumably from HNO\textsubscript{4} and MPN (even though a correction for MPN had already been applied). In DC3 convective outflow HNO\textsubscript{4} was typically 1%–2% of NO\textsubscript{2}, suggesting that our LNO\textsubscript{3} PE could be biased high by a very small percentage due to this interference. Reed et al. (2016) indicate that 5% of PAN decomposes in the photolytic conversion of NO\textsubscript{2} to NO prior to chemiluminescent detection, leading to a small high bias in NO\textsubscript{2} observations. At the PAN and NO\textsubscript{2} mixing ratios found in the 29–30 May storm anvil, this high bias would have been 1.25%–2.5% in the peak periods of NO\textsubscript{2} due to lightning and 3%–6% elsewhere. Overall, we consider 82 mol fl\textsuperscript{-1} as a lower bound for the PE of this storm and believe the true value lies in the 80–110 mol fl\textsuperscript{-1} range.

5.2. Downwind Chemistry

5.2.1. Downwind Observations Over the Southern Appalachians

Based on a combination of near-real-time GOME-2 satellite NO\textsubscript{2} observations and forward trajectories, the GV and DC-8 were directed toward the southern Appalachian region on 30 May 2012 to locate and sample the convective outflow plume associated with the severe supercell system that developed along the Oklahoma/Kansas border at 21:10 UTC 29 May and moved east-southeast through Oklahoma. The aircraft set up a triangular shaped flight path from Lynchburg, Virginia, to Athens, Georgia, to Snowbird (a waypoint on the Tennessee/North Carolina border). Although downwind sampling was conducted from 7.5 to 15.5 km in this region, most of the simulation. The loss due to formation of HNO\textsubscript{2} and organic nitrates can be estimated at 0.4 ppbv by subtracting the “observed LNO\textsubscript{3}” (0.8 ppbv) from the passive LNO\textsubscript{3} (1.2 ppbv) obtained in this emission scenario. The results in Table 4 for this and other scenarios also suggest that there is NO\textsubscript{3} loss within the cloud due to chemistry (i.e., formation of HNO\textsubscript{2} and organic nitrates). For the 82 mol flash\textsuperscript{-1} scenario, the mean total NO\textsubscript{3} with lightning (1.3 ppbv) is less than the sum of the average model-simulated LNO\textsubscript{3} (1.2 ppbv) within the anvil outflow and the mean NO\textsubscript{3} from the WRF-Chem simulation with no lightning (0.6 ppbv).

In the model simulation, NO\textsubscript{3} and LNO\textsubscript{3} maxima were co-located with the O\textsubscript{3} minimum in the upper portion of the convective core (> 7 km), as well as the anvil. Here the mixing ratios for NO\textsubscript{3} and LNO\textsubscript{3} exceed 2 ppbv. While vertical cross-sections suggest a small amount of boundary layer NO\textsubscript{3} is transported into the upper region of the convective cells, the model-simulated LNO\textsubscript{3} appears to make up 88% of the NO\textsubscript{3} within the anvil.

The LNO\textsubscript{3} PE values obtained in this study were obtained by tuning the LNO\textsubscript{3} source to ensure that model values match observations. Thus, incomplete model chemistry and/or measurement interferences add uncertainty to the results. Nault et al. (2016, 2017) found that the lifetime of NO\textsubscript{3} in the near field of deep convection is much shorter than had been assumed. They indicate that the lifetime may be 3 hr in the near-field outflow from thunderstorms due to rapid production of methyl peroxy nitrate (MPN) and alkyl nitrates, but the lifetime becomes much longer as the air moves downwind. The MOZART-4 mechanism does not contain MPN, and production of alkyl nitrates is not well represented. These findings suggest that our model-based estimate of LNO\textsubscript{3} PE may be biased low because of a lack of adequate accounting for these nitrate compounds, leading to a need for a resulting larger value of LNO\textsubscript{3} production per flash. Nault et al. (2017) indicate that models without the revised chemistry mentioned above may underestimate LNO\textsubscript{3} PE by up to 33%. Brune et al. (2021) found large enhancements of OH and HO\textsubscript{2} in DC3 storms that were linked with visible and subvisible lightning flashes. The lightning-enhanced HO\textsubscript{2}\textsubscript{(LNO\textsubscript{3})} leads to increased HONO production (a few pptv in the presence of the 1–2 ppbv LNO\textsubscript{3} observed in this case). Again, our model LNO\textsubscript{3} production calculation could have a small low bias due to unaccounted HONO production. Shah et al. (2023) concluded that there was interference in the DC3 TD-LIF measurements of NO\textsubscript{2}, presumably from
of the DC-8 sampling was at an altitude of 11 km from 21:00 UTC 30 May to 00:00 UTC 31 May, and most of the GV sampling was at altitude of 11.4–11.5 km from 22:00 UTC to 23:30 UTC 30 May.

For the downwind trace gas analyses, the 1-sec DC-8 and GV aircraft data were averaged over 5 and 4 s time intervals, respectively, to represent a 1 km horizontal resolution based on the aircraft ground speeds. In addition, only observations within tropospheric air were included (O$: CO$ ratio <1.25; Hudman et al., 2007). Plots of the trace gas mixing ratios along the aircraft flight tracks suggest that the aircraft encountered the outflow plume along the southwest-to-northeast tracks between Athens and Lynchburg. Figures S1 and S2 in Supporting Information S1 show aircraft detected NO$\textsubscript{2}$ mixing ratios of 1–2 ppbv at 10–12 km altitude along the southern track and the northeast corner of the triangle near Lynchburg. The CO measurements are generally above 110 ppbv within the sampling region (Figures S3 and S4 in Supporting Information S1); however, the portion of the track where NO$\textsubscript{2}$ is elevated coincides with slightly higher CO mixing ratios (120 ppbv). The aircraft observations suggest that the aircraft did encounter the outflow plume from the previous afternoon/evening, as indicated by the collocation of the highest NO$\textsubscript{2}$ and CO mixing ratios. The DC-8 observed enhanced O$\textsubscript{3}$ (110–130 ppbv) between 10.5 and 12.0 km altitude in several locations in the western half of the triangle and as it approached and exited the area (Figure S5 in Supporting Information S1). The GV also found O$\textsubscript{3}$ mixing ratios between 110 and 130 ppbv; however, these measurements were observed between about 11.0 and 12.5 km altitude along the northern portion of the sampling region (Figure S6 in Supporting Information S1). The flight tracks appear to show that the areas with higher O$\textsubscript{3}$ mixing ratios (110–130 ppbv) occurred where the NO$\textsubscript{2}$ < 1 ppbv. Elevated NO$\textsubscript{2}$ (>1 ppbv) occurred in locations where O$\textsubscript{3}$ mixing ratios were smaller (80–110 ppbv), suggestive of convective outflow.

Backward trajectories from the NOAA HYSPLIT model (NOAA, 2023; Rolph et al., 2017; Stein et al., 2015) indicated that at 18:00 UTC on 30 May the air masses at 8, 10, and 12 km altitude within the region of enhanced LNO$\textsubscript{2}$ passed through lightning activity over the previous 12–42 hr Figure 11a shows the 12 km trajectory passed through the area of Oklahoma where convection was sampled by the DC3 aircraft between 18 UTC 29 May and 00 UTC 30 May, the 10 km trajectory passed through convection in the same time period in southern
Figure 12. Mean trace gas mixing ratios based on DC-8 and GV observations taken over the southern Appalachian region within tropospheric air on 30 May 2012. Each horizontal line represents 1 standard deviation.

Oklahoma, and the 8 km trajectory intercepted lightning at 00 UTC 29 May, which was farther upstream and not part of the previous day’s research flight. Figure 11b indicates that the 10 and 12 km trajectories passed near the supercell system between 00 and 06 UTC 30 May. Other points on the south end of the sampling region may have had some influence of convection in Mississippi/Alabama overnight. The vast majority of the downwind impact on NO₂ was from storm systems in Oklahoma on the previous day.

Figure 12 shows the mean mixing ratio profiles within tropospheric air for five trace gases measured by the aircraft over the southern Appalachian region. The CO and NO₂ profiles suggest the outflow plume may extend from roughly 8–12 km; however, the mean NO₂ profile appears to show the core of the plume is located between 10 and 12 km, as suggested by the back trajectories. When the aircraft sampled the anvil outflow of the active convection the previous evening, the average O₃ mixing ratio between 10.7 and 12.2 km altitude was 86 ppbv. Based on the mean observed O₃ profile from the downwind region in Figure 12, the average O₃ mixing ratio within this same altitude range is 105 ppbv, indicating a mean increase of 19 ppbv with increases at some altitudes of 25–30 ppbv. We use the 36-km horizontal resolution WRF-Chem simulation to estimate how much of the increase in O₃ at the primary DC-8 and GV altitudes is due to photochemical production in the 12 hr of sunlight that the convective outflow plume experienced during transport to the location of the beginning of downwind sampling.

5.2.2. Regional Model Simulation Results

The DC-8 and GV ended their sampling of the active convection in Oklahoma at approximately 01:00 UTC on 30 May. We compare the mean vertical profile of in-cloud ozone observed along the aircraft measurement tracks with that from the 36-km horizontal resolution WRF-Chem simulation in Figure 13. We removed the points affected by stratospheric ozone for both aircraft measurements and simulations of the cloud outflow by excluding the points where the observed and modeled ratio of O₃ to CO was greater than 1.25. It is not appropriate to include these points as the model does not include a full representation of stratospheric chemistry. Therefore, as a result there were no remaining model simulation results to plot on the level of 12.25 km in Figure 13. However, for the
other levels the model well reproduced the mean ozone vertical distribution. The aircraft and simulation mean ozone mixing ratios in the storm anvil outflow region are shown in Table 5. The GV measured mean ozone mixing ratio was 82.9 ppbv and the DC-8 measured mean ozone mixing ratio was 78.1 ppbv. The model simulated mean ozone mixing ratio at GV measuring altitudes was 81.6, and the simulated average ozone mixing ratio at DC-8 measuring altitudes was 79.8 ppbv. The model simulated mean ozone mixing ratios in the storm were close to the observations (the differences between the observation and simulation were less than 2%). Note the large reduction in O$_3$ from the initial condition profile that resulted from convective transport of low O$_3$ from the boundary layer.

The hourly simulated CO, NO, and O$_3$ results show the progression of the storm outflow from Oklahoma to the southern Appalachian region (Figures S7–S15 in Supporting Information S1) at the mean downwind DC-8 measurement altitude (11 km, left) and at the mean GV altitude (11.5 km, right). Figure 14 shows the model simulated NO and O$_3$ fields for 11 km at the approximate time of the storm dissipation (04:00 UTC). A very prominent NO$_x$ maximum and O$_3$ minimum appear in northeastern Oklahoma at this time. NO$_x$ exceeded 2 ppbv over a large area with a peak mixing ratio just over 5 ppbv, while the O$_3$ minimum over roughly the same area was 60–70 ppbv. Note the significant stratospheric intrusion evident in the O$_3$ field at this time to the east of the convective outflow. Figure 15 shows the model simulated NO and O$_3$ fields again at 11 km compared with the aircraft measurements over the hours 21:00 UTC 30 May to 00:00 UTC 31 May. As the modeled storm started 80 min later than the observed system, we plotted the downwind aircraft data from the hour 80–140 min earlier in these figures than the model time. The model reproduced the observed upper tropospheric NO$_x$ and ozone downwind horizontal transport. The high CO (see Figures S7–S9 in Supporting Information S1). NO$_x$ and low O$_3$ upper tropospheric air, which represented the storm downwind outflow, was transported to the southern Appalachian region by late afternoon on 30 May. At 21:00 UTC the model shows an area of enhanced NO$_x$ (1–2 ppbv) stretching from southern West Virginia across southwestern Virginia and into eastern Tennessee. This NO$_x$ enhancement moved slowly eastward over the next 3 hours. An O$_3$ minimum in the model output still persisted over these 4 hr, now located over eastern Tennessee, western North Carolina and northern Georgia. However, it was nowhere near as pronounced as at 04:00 UTC likely as a result of photochemical production of O$_3$ during the sunlit hours of 30 May. The model shows remnants of the stratospheric intrusion to the east of the O$_3$ minimum, although this does not agree with the DC-8 observations of O$_3$ near 23:00 UTC (see Figure 15 and Figures S13–S15 in Supporting Information S1).

It appears that the model eastward advection was too slow. The peak DC-8 observed CO was also farther east than the model peak at 23:20 and 00:20 UTC (Figure S9 in Supporting Information S1), also suggesting eastward advection in the model was not as fast as observed. Dilution of the CO plume in the model also appears to be greater than observed.

Table 5

<table>
<thead>
<tr>
<th>Aircraft-Measured and Model-Simulated Mean Ozone (ppbv) in the Storm Anvil, One Day Downwind of Storm (ppbv), Net Ozone Change (ppbv), and Simulated Net Ozone Production (ppbv Day$^{-1}$)</th>
<th>DC-8 measuring altitude</th>
<th>GV measuring altitude</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>OBS</td>
<td>WRF-chem</td>
</tr>
<tr>
<td>Storm region</td>
<td>78.1</td>
<td>5.3</td>
</tr>
<tr>
<td>1 day after</td>
<td>104.3</td>
<td>3.3</td>
</tr>
<tr>
<td>Net ozone change</td>
<td>26.2</td>
<td>17.3</td>
</tr>
<tr>
<td>Net ozone production</td>
<td>–</td>
<td>11.0</td>
</tr>
</tbody>
</table>
Figure 14. WRF-Chem simulated NO\textsubscript{x} (upper) and O\textsubscript{3} (lower) at 36-km horizontal resolution for 11 km at 04:00 UTC 30 May 2012.

Figure 15. Similar to Figure 14, except for 21:00, 22:00, 23:00 UTC 30 May 2012 and 00:00 UTC 31 May 2012. NO\textsubscript{x} (left) and O\textsubscript{3} (right).
The aircraft measured ozone on 30 May in the downwind region was larger than the ozone mixing ratio measured in the anvil outflow during the active storm (Table 5). The mean ozone mixing ratio at 11 km measured by the DC-8 was 104.3 ppbv, which was 26.2 ppbv larger than one day before in the anvil outflow, while the mean ozone mixing ratio at 11.4 km measured by the GV was 113.5 ppbv, which was 30.6 ppbv larger than one day before. We calculated the simulated mean ozone mixing ratio in the downwind outflow region at 00:00 UTC 31 May along the aircraft flight track. The storm downwind outflow region was defined as the region where NO$_3$ > 0.5 ppbv. The simulated mean ozone mixing ratios in that region were similar to the observations (Table 5). The simulated mean ozone mixing ratio at 11 km was 96.4 ppbv (7.6% lower than the observation), and 115.8 ppbv (2% higher than the observation) at 11.4 km. Therefore, the simulated average ozone increase from the in-storm observations was 17.3 ppbv at the mean DC-8 measuring altitude and 34.2 ppbv at the mean GV altitude. The simulated increase was close (11%) to the observed increase at 11.5 km, but 34% smaller than observed at 11 km.

Not all of the O$_3$ increases were caused by photochemical production. Some of the increase was likely due to mixing of larger ozone values (e.g., the stratospheric intrusion of large ozone values out ahead of the convective outflow plume) from areas surrounding the low ozone plume as it moved downwind. In Figure S13 in Supporting Information S1, before sunrise time (approximately 10:30 UTC on 30 May), the low ozone region started to shrink between 08:00 UTC and 10:00 UTC at the GV altitude, which suggests that the ozone increase was due to mixing during this period. After sunrise, ozone rapidly increased in this air mass due to photochemical production. The ozone production results are shown in Figure 16 and Table 5. The mean simulated net ozone production was 11.0 ppbv day$^{-1}$ (64% of the total simulated ozone change) at the mean DC-8 altitude (11 km), and 5.5 ppbv day$^{-1}$ (16% of the total simulated ozone change) at the mean GV altitude (11.4 km). Thus, at the DC-8 altitude the net ozone production dominated the ozone change, while at the GV altitude, the mixing with larger ozone values dominated the ozone change. At altitudes between 9 and 11 km, where NO$_3$ was also enhanced in the storm outflow, net ozone production in the model proceeded at a rate of between 10.1 and 10.8 ppbv day$^{-1}$. At 10.5 km the net ozone production (10.1 ppbv day$^{-1}$) accounted for an even larger fraction (83%) of the simulated total ozone change (12.1 ppbv day$^{-1}$) over 24 hr. From 9 to 10 km net ozone production was 10.4–10.8 ppbv day$^{-1}$, but mixing of lower ozone air into the outflow plume during transport led to simulated total ozone changes over 24 hr from 7 to 5 ppbv at these levels.

6. Discussion

6.1. Comparison of Cloud-Resolved LNO$_3$ Production Results With Previous Work

Schumann and Huntrieser (2007) obtained 250 mol flash$^{-1}$ (range of 33–660 mol flash$^{-1}$) as a best estimate of LNO$_3$ PE based on their review of theoretical considerations, laboratory studies, field data, cloud-resolved modeling, and other studies from 1976 to 2007. Estimates of LNO$_3$ production deduced from cloud-resolved modeling constrained by anvil NO$_3$ observations have come from simulations of storms in several field
Figure 17. Correlation between the observed 10-min mean flash rate and (top) mean flash extent and (bottom) mean energy per flash within the coverage of the central OK lightning mapping array network for the 29 May case (23:40–03:00 UTC). The red plus represents the data points within the 3D coverage. The purple asterisk represents the data points prior to the 3D coverage (21:30–23:30 UTC). The blue cross represents the data points after the 3D coverage (03:10–04:20 UTC). The fitted lines and correlations are for only the 3-D coverage points.

Evidence from field campaigns has shown a relationship between flash rate and flash size. In a study of two 2004 supercells, Bruning and MacGorman (2013) reported that smaller flash extents were associated with higher average flash rates. Similar associations were noted by Carey et al. (2005), Kuhlman et al. (2006), and Weiss et al. (2012), as well as studies from the DC3 campaign (Bruning & Thomas, 2015; Carey et al., 2019; Mecikaliski et al., 2015). Smaller flashes (less channel length) are associated with less NO production (Carey et al., 2016). Data from the Oklahoma LMA for the 29–30 May storm lend support for these relationships. Figure 17 shows the LMA data for flash extent (estimated as the square root of the flash area based on the convex hull formed by the sources) plotted versus that for flash rate. For 10-min periods when the storm was within the 3-D detection region of the LMA there is a negative correlation, with decreasing flash extent associated with greater flash rates. This tendency was most evident at flash rates from 0 to 100 flashes per 10 min. No clear relationship was noted when the storm was in the 2-D detection region, where the LMA flash extent data may be of poorer quality. Similarly, a negative correlation of flash energy and flash rate was noted for the 3-D detection period. Flash energy is assumed to be proportional to flash area and estimated as in Bruning and Thomas (2015). Flash energy has been related to LNO₃ production in theoretical considerations and in laboratory experiments (e.g., Chameides et al., 1977). Therefore, the LMA data suggest less LNO₃ production per flash at high flash rates, such as in the 29–30 May Oklahoma storm, for which we estimate a relatively low overall mean production rate of 80–110 mol flash⁻¹. However, given the range of 10-min mean values of flash extent and flash energy noted in Figure 17, the LNO₃ production rate per flash had significant variability over the time period of the 3-D LMA data.

Other DC3 analyses and modeling have yielded estimates of LNO₃ production per flash. Pollack et al. (2016) performed analyses of the NOₓ data observed from the aircraft transects of six DC3 storms. They used a combination of observations of storm inflow and outflow from all three aircraft, spatial information from ground-based radars and satellite observations, and intracloud and cloud-to-ground lightning flashes from the LMA. Two analysis methods (a volume-based approach and a flux-based approach) were used for converting enhancements in lightning-produced NOₓ to moles NOₓ per flash. Results from the more robust volume-based approach for three storms sampled over Oklahoma and Colorado for which there is the greatest confidence suggest a range of 142–291 (average of 194) mol flash⁻¹ (or 117–332 mol flash⁻¹ including uncertainties). As mentioned in Section 5.1.2, Pollack et al. (2016) found a value of 10⁷ ± 24 mol flash⁻¹ for the 29–30 May storm, but suspected that the storm volume used in the calculation may have been biased low. Our value of 80–110 mol per flash based on our model simulations encompasses the Pollack et al. value for this case, suggesting that the Pollack value may not have a significant bias. Our model simulation approach provides important validation of the Pollack et al. work. The model provides a more comprehensive view of LNO₃ in the anvil than solely the aircraft observations, which cover only a short period of time and a small fraction of the storm volume. Figure 18 shows the relationship between mean LNO₃ production for the Pollack et al. storms and mean flash area from the LMA data. Note that here we use mean flash area, whereas total (cumulative) flash area is used in a similar plot in Pollack et al. The data point representing the results from our simulation for the 29–30 May storm is also included. A moderate positive correlation of 0.42 was found.

Carey et al. (2016) used the Lightning Nitrogen Oxides Model (LNOM) of Koschak et al. (2014) to estimate LNO₃ production in the 21 May 2012 multi-cell storm observed in Alabama during DC3. The LNOM assumes that the
LNO$_2$ production per unit flash channel length is much larger in a CG flash than in an IC flash. They obtained 919 mol per flash for CG flashes and 116 mol per flash for IC flashes. Despite the summed IC flash extent being much larger than the summed CG flash extent, the majority of LNO$_2$ production was estimated to be from CG flashes. However, these LNO$_2$ production values were not constrained by DC3 aircraft NO$_2$ observations. Davis et al. (2019) used gridded flash extent data from the Colorado and Alabama LMAIs to estimate LNO$_2$ production using the formulation of Wang et al. (1998), which utilizes the pressure at which the flash channel is located. Their findings were that LNO$_2$ production per flash in Colorado anomalous polarity storms (mean of 124 mol flash$^{-1}$) was greater than in Alabama normal polarity storms (77 mol flash$^{-1}$). The LNO$_2$ production uncertainties associated with different analysis methods were illustrated by Pollack et al. (2016). Analyzing a June storm over Colorado, Davis et al. obtained a PE of 190 mol flash$^{-1}$ using a flux-based approach and 72 mol flash$^{-1}$ using LMA flash extent data. They also found that the flash extent (and LNO$_2$ production) maximized at a lower altitude in the anomalous polarity storms than in the normal polarity storms. Our estimate of 80–110 mol flash$^{-1}$ for the 29–30 May Oklahoma storm from our cloud-resolved modeling approach is close to the Davis et al. value for the anomalous polarity Colorado storm. However, with the elevated (9–11 km) maximum of flash extent, the Oklahoma storm was likely of normal polarity.

Satellite and airborne remote sensing of NO$_2$ has allowed estimates of LNO$_2$ over large numbers of storm systems. Special retrievals have been conducted to best obtain the LNO$_2$ signal. This procedure has involved use of an air mass factor appropriate for the vertical profiles of NO and NO$_2$ resulting from lightning in a storm in order to convert slant columns of NO$_2$ to vertical columns of NO$_2$. Laughner and Cohen (2017) showed that the Ozone Monitoring Instrument (OMI) on the Aura satellite has high sensitivity to NO$_2$ in the upper troposphere resulting from lightning during DC3. Pickering et al. (2016) used five summers (2007–2011) of NO$_2$ data from OMI along with flash data from the World Wide Lightning Location Network (WWLLN) over the Gulf of Mexico to obtain a mean value of LNO$_2$ production per flash of 80–45 mol per flash. This range of values encompasses the results we obtained for the 29–30 May DC-3 storm. Buscela et al. (2019) also used the OMI and WWLLN data for the same five summers in a gridded LNO$_2$ analysis over the Northern Hemispheric mid-latitude continental regions. The resulting mean LNO$_2$ production was 180–100 mol per flash, also encompassing the 29–30 May storm results. Buscela et al. (2019) also found that a power law best fit the relationship between LNO$_2$ production per flash and flash rate, with the per flash production lower in storms with more frequent flashes. This result is consistent with our results for the high flash rate 29–30 May storm. D. J. Allen, Pickering, Lamsal, et al. (2021) found somewhat larger LNO$_2$ production over 10 US and western Atlantic storms observed by the GeoCAPE Airborne Spectrometer (GCAS) on the NASA ER-2 aircraft during Spring 2017. D. J. Allen, Pickering, Bucseca, et al. (2021) used Tropospheric Monitoring Instrument (Tropomi) NO$_2$ data along with flashes from the Geostationary Lightning Mappers (GLM) on the GOES-16 and -17 satellites and from the ENTLN for 29 US storms, obtaining 175–100 mol/flash for GLM and 120–65 mol/flash for ENTLN. The LNO$_2$ production range for the 29–30 May storm is well within these ranges. In both the GCAS and Tropomi work, the LNO$_2$ production per flash was negatively correlated with flash density. Therefore, the 29–30 May results are in agreement with the ranges of LNO$_2$ production per flash determined from remote sensing, and corroborate the finding of relatively low production per flash in high flash rate storms.

### 6.2. Comparison of Downwind Ozone Production Results With Previous Work

Prior model simulations have generally shown O$_3$ enhancements of similar magnitude downwind of convection as we have computed for the 29–30 May Oklahoma storm. DeCaria et al. (2005) found O$_3$ increased on average by 10 ppbv day$^{-1}$ following a cloud-resolved simulation of the 12 July 1996 storm observed in Colorado during the STERAO-A campaign. Ott et al. (2007) simulated a storm in the high NO$_x$ environment near Munich, Germany during EULINOX. Simulations of the chemical environment in the free troposphere over the 24 hr following the storm showed on average a small increase in the net production of ozone at most levels resulting from LNO$_2$. 

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**Figure 18.** Correlation between the mean flash area and mean LNO$_2$ production per flash (from Pollack et al., 2016) for six convective events observed during DC3. The squares and triangles represent Oklahoma and Colorado convection, respectively. LNO$_2$ production using the flux-based approach is represented by orange and blue, while the volume-based approach is represented by red and purple. The green asterisk represents the 29–30 May event based on data within the 10-min moving spatial masks and 3D coverage of the central OK lightning mapping array network (23:40–03:00 UTC). Fitted line and correlation are for all points plotted. Results suggest mean P(NO$_2$) per flash is ∼100 mol flash$^{-1}$ for very small flashes.
maximizing at approximately 5 ppbv per day at 5.5 km. Other estimates include those by Ren et al. (2008) downwind of frequent convection over the US (11 ppbv per day) and more rapid production in outflow enriched with both LNO$_2$ and isoprene (10 ppbv in a few hours; Apel et al., 2012). Box model analyses by Apel et al. (2015) showed O$_3$ increased downwind of two thunderstorms observed on 22 June 2012 in Colorado during DC3 by 11–14 ppbv over a two-day period. More rapid net ozone production (13–14 ppbv over 5 hr) was noted by Brune et al. (2018) in the outflow from the 21 June 2012 MCS. Compared to the net ozone production results from the previous studies, our simulated net ozone production at the DC-8 altitude was within the range of most of these estimates, while our net ozone production at the GV altitude was approximately equal to or smaller than the results from most previous studies.

The O$_3$ enhancement observed and simulated in convective outflow is greater than in the clean free troposphere not only because of enhanced NO$_x$, but also because of increased HO$_x$ and HO$_2$ precursors in the outflow plume. Updrafts from the active convection the previous afternoon/evening transported boundary layer air, which contained HO$_x$ precursors such as HCHO, H$_2$O$_2$, and CH$_3$OOH, into the upper convective region of the Oklahoma thunderstorms (see Bela et al., 2016, 2018). Despite loss due to wet scavenging, these gases were still enhanced over background as the air was detrained from the 29–30 May storm. Our downwind model simulation indicates that mixing of higher ozone air with a stratospheric origin into the convective outflow was responsible for a substantial portion of the increase in ozone in the upper portion of the convective outflow reaching the downwind region (36% at 11 km and 84% at 11.4 km). Based on an analysis of DC3 aircraft observations, Schroeder et al. (2014) suggest the mixing of polluted convective outflow and stratospheric air rapidly produces OH. Three of the stratospherically influenced samples in the Schroeder et al. (2014) analysis were from the 29–30 May convective event. The presence of LNO$_2$ and boundary layer trace gases aided in producing more O$_3$ in the downwind plume on 30 May than would have been present in the background air. Indeed, at 11 km and below photochemical production of O$_3$ dominated the O$_3$ increase. While convective transport of the low O$_3$ air from the PBL substantially reduced O$_3$ in the upper tropospheric outflow, photochemical production of O$_3$ during downwind transport resulting from enhanced NO$_x$ due to lightning played a major role in restoring upper tropospheric O$_3$ to nearly its magnitude prior to the storm.

7. Summary

Using the ICEFLUX FRPS scheme, several WRF-Chem simulations were run to investigate the mean LNO$_2$ production per flash and the transport and distribution of other trace gases for the 29–30 May 2012 convective event. The simulations included one run with no lightning and seven runs with different LNO$_2$ production scenarios. A comparison of the aircraft and model-simulated CO and O$_3$ mixing ratios indicates that the convective transport is slightly underestimated in the model. To compensate for the transport underestimate, one model layer below each layer the aircraft flew within was analyzed. This adjustment showed the model-simulated CO and O$_3$ mixing ratios are within 1% and 3% of the aircraft observations, respectively.

With lightning chemistry turned off in the model, the overall mean NO$_x$ from the four model layers the aircraft flew in is underestimated by 55%. When observed IC:CG ratios and vertical distributions of flash channel segments are incorporated in the model, the 125 mol per flash scenario from Bela et al. (2016) overestimated NO$_x$ in the anvil by 35%. A comparison of the aircraft and model-simulated NO$_x$ mixing ratios from six additional LNO$_2$ production scenarios suggested 82 mol flash$^{-1}$ achieved a best match within 7% of the observations. The analyzed storm is a high flash rate event with small flash extents (6.5 km), which suggests that LNO$_2$ production per flash would be low. However, 82 mol flash$^{-1}$ is likely a lower bound, as some NO$_x$ species are either underestimated or not represented in the model. Consideration of chemistry aspects of DC3 storms found by other investigators (Brune et al., 2021; Nault et al., 2017) leads us to our best estimate of 80–110 mol fl$^{-1}$ for the 29–30 May storm. The midpoint of this estimate is roughly 2.6 times smaller than the mean 250 mol NO flash$^{-1}$ calculated by Schumann and Huntrieser (2007) from a survey of the literature which included theoretical estimates, and values derived from laboratory studies, field data, and cloud-resolved model simulations from 1976 to 2007; however, the value derived for the 29–30 May DC3 storm is within the lower end of the range of estimates (33–660 mol flash$^{-1}$) reported by Schumann and Huntrieser. The estimate for the observed storm is also 5 times smaller than the mean value suggested by Ott et al. (2010) for midlatitude and subtropical storms (500 mol flash$^{-1}$). Our results are in agreement with the findings of Pollack et al. (2016), who showed that this high flash rate storm had a relatively low mean LNO$_2$ production per flash, thereby validating their results.
A weak positive correlation was found between LNO₃ production and the total flash area over several DC3 storms (Pollack et al., 2016). The analysis here of a more meaningful relationship between mean LNO₃ production per flash and mean flash area over the several storms (including the 29–30 May case) yielded a more moderate positive correlation. When combined with the relative negative correlation found between the total flash rate in the observed 29–30 May severe supercell system and flash extent ($r = 0.69$) or energy per flash ($r = 0.70$), it is possible that a new parameterization scheme may be developed in the future to estimate LNO₃ production per flash as a function of time from the flash rate in models.

One of the goals in DC3 was to quantify changes in the composition and chemistry of the upper troposphere downwind of deep convection by taking measurements of the convective outflow while the storm is active and again 12–48 hr downwind. Based on aircraft observations, there was a mean 19 ppbv increase of O₃ over the 8–12 km layer during the outflow plume’s transit to the southern Appalachian region. The 36-km horizontal resolution model well reproduces the convective transport of ozone and the downwind ozone transport and production. When the storm was active, the upper tropospheric ozone mixing ratio was reduced by 20–40 ppbv due to the vertical transport of low ozone PBL air. During the nighttime hours the ozone in the upper tropospheric outflow continued to decrease. After sunrise, the ozone mixing ratio increased rapidly, which resulted from the photochemical reactions. The aircraft measured mean ozone mixing ratio increased 26.2 ppbv at the mean DC-8 altitude, and 30.6 ppbv at the mean GV altitude in the downwind outflow region compared with the anvil region. The average ozone increase in the model was 17.3 ppbv at the primary DC-8 altitude and 34.2 ppbv at the primary GV level. The simulated net ozone production was 11.0 ppbv day⁻¹ (64% of the total simulated ozone increase) at the mean DC-8 altitude, and 5.5 ppbv day⁻¹ (16% of the total simulated ozone increase) at the mean GV altitude. Thus, at the DC-8 altitude the net ozone production dominated the ozone change, while at the GV altitude, the mixing of larger ozone values dominated the ozone change. At 10.5 km the net production accounted for 83% of the total ozone change over the downwind transport. Compared to the net ozone production results from the previous studies, the simulated net ozone production at DC-8 altitude was within the range, while the net ozone production at GV altitude was approximately equal to or lower than the results from the previous studies. Photochemical production of O₃ resulting from LNO₃ had a major role in restoring upper tropospheric O₃ in these air parcels to nearly its magnitude prior to the Oklahoma storm.

**Data Availability Statement**

The WRF-Chem version 3.6.1 code is available at https://www2.mmm.ucar.edu/wrf/users/download/get_source.html and the modified WRF-Chem source code for our simulations is available at https://dsrs.atmos.umd.edu/DATA/WRF-ChemV3.6.1-modified-scripts/. Flash statistics from the Oklahoma LMA are available at https://data.eol.ucar.edu/dataset/353.201. The DC3 and GV merged aircraft data are available at the NASA data repository for Airborne Science Data for Atmospheric Composition website (https://www-air.larc.nasa.gov/cgi-bin/Avl2v3/komine/dc3). NAM-ANL is available from https://www.ncei.noaa.gov/products/weather-climate-models/north-american-mesoscale. MOZART-4 global model output is no longer available from NCAR for use as WRF-Chem initial and boundary conditions. However, NCAR now recommends use of CAM-Chem data (https://www2.acom.ucar.edu/gcm/cam-chem-output) for this purpose. The NOAA HYPLIT model is available at http://www.ready.noaa.gov/HYPLIT.php. NEXRAD data for the DC3 storm are available from http://catalog.eol.ucar.edu/cgi-bin/dc3_2012/radar/index. GOES-13 satellite products are available from http://catalog.eol.ucar.edu/cgi-bin/dc3_2012/ops/index. NLDN data may be purchased from Vaisala. ENTLN data may be freely requested from Earth Networks, Inc. (Email: info@earthnetworks.com). WRF-Chem model output are stored at the following locations: https://dsrs.atmos.umd.edu/DATA/1km_simulation/ and https://dsrs.atmos.umd.edu/DATA/36km_simulation/.

**References**


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