A cloud-scale model study of lightning-generated NO\textsubscript{x} in an individual thunderstorm during STERAO-A

Alex J. DeCaria, Kenneth E. Pickering, Georgiy L. Stenchikov, John R. Scala, Jeffrey L. Stith, James E. Dye, Brian A. Ridley, and Pierre Laroche

Abstract. Understanding lightning NO\textsubscript{x} (NO + NO\textsubscript{2}) production on the cloud scale is key for developing better parameterizations of lightning NO\textsubscript{x} for use in regional and global chemical transport models. This paper attempts to further the understanding of lightning NO\textsubscript{x} production on the cloud scale using a cloud model simulation of an observed thunderstorm. Objectives are (1) to infer from the model simulations and in situ measurements the relative production rates of NO\textsubscript{x} by cloud-to-ground (CG) and intracloud (IC) lightning for the storm; (2) to assess the relative contributions in the storm anvil of convective transport of NO\textsubscript{x} from the boundary layer and NO\textsubscript{x} production by lightning; and (3) to simulate the effects of the lightning-generated NO\textsubscript{x} on subsequent photochemical ozone production. We use a two-dimensional cloud model that includes a parameterized source of lightning-generated NO\textsubscript{x} to study the production and advection of NO\textsubscript{x} associated with a developing northeast Colorado thunderstorm observed on July 12, 1996, during the Stratosphere-Troposphere Experiment—Radiation, Aerosols, Ozone (STERAO-A) field campaign. Model results are compared with the sum of NO\textsubscript{x} measurements taken by aircraft and photostationary state estimates of NO\textsubscript{2} in and around the anvil of the thunderstorm. The results show that IC lightning was the dominant source of NO\textsubscript{x} in this thunderstorm. We estimate from our simulations that the NO\textsubscript{x} production per CG flash (P\textsubscript{CG}) was of the order of 200 to 500 mol flash\textsuperscript{-1}. NO\textsubscript{x} production per IC flash (P\textsubscript{IC}) appeared to be half or more of that for a CG flash, a higher ratio of P\textsubscript{IC}/P\textsubscript{CG} than is commonly assumed. The results also indicate that the majority of NO\textsubscript{x} (greater than 80%) in the anvil region of this storm resulted from lightning as opposed to transport from the boundary layer. The effect of the lightning NO\textsubscript{x} on subsequent photochemical ozone production was assessed using a column chemical model initialized with values of NO\textsubscript{x}, O\textsubscript{3}, and hydrocarbons taken from a horizontally averaged vertical profile through the anvil of the simulated storm. The lightning NO\textsubscript{x} increased simulated ozone production rates by a maximum of over 7 ppbv d\textsuperscript{-1} in the upper troposphere downwind of this storm.

1. Introduction

The nitrogen oxides NO and NO\textsubscript{2} (collectively designated as NO\textsubscript{x}) are included in global chemical transport models (CTMs) because they are important catalysts for the production of tropospheric ozone and are involved in numerous other reactions in the atmosphere. Lightning may be a particularly significant contributor to NO\textsubscript{x} mixing ratios in the upper troposphere [Ridley et al., 1996]. A key for improving the parameterization of lightning NO\textsubscript{x} production in global-scale models is an understanding of the phenomena on the scale of individual thunderstorms.

Of all the sources of tropospheric NO\textsubscript{x}, the one with the greatest uncertainty is that due to lightning [Lawrence et al., 1995; Price et al., 1997]. The uncertainty has been reduced over the past decade, with most recent studies yielding values of from 2–20 Tg(N) yr\textsuperscript{-1}; however, there is still disagreement over whether the value is likely to be at the low or high end of this range. Three recent global studies illustrate the ongoing uncertainty, with Price et al. [1997] suggesting a value of 12.2 Tg(N) yr\textsuperscript{-1} and stating that it is extremely unlikely that the value is less than 5 or greater than 20 Tg(N) yr\textsuperscript{-1}, while Levy et al. [1996] gave a most probable range of 3–5 Tg(N) yr\textsuperscript{-1}, and Wang et al. [1998a] gave a value of 3 Tg(N) yr\textsuperscript{-1}. The disparity is due to uncertainty in both the average global frequency of lightning and the average production of NO\textsubscript{x} per lightning flash. Compounding the problem is that intracloud (IC) and cloud-to-ground (CG) flashes may have different values of NO\textsubscript{x} production per flash. The production of NO\textsubscript{x} from an IC flash (P\textsubscript{IC}) is usually scaled to that from a CG flash (P\textsubscript{CG}). Widely differing values for the ratio P\textsubscript{IC}/P\textsubscript{CG} are used in the literature; for example, Biazar and McNider [1995] assumed P\textsubscript{IC}/P\textsubscript{CG} to be near zero, Price et al. [1997] used a value of 0.1,
while Wang et al. [1998a] used a value of 0.3. Gallardo and Cooray [1996] suggested that the ratio might even be close to unity.

Cloud-scale field studies of NO\textsubscript{x} in thunderstorms were first extrapolated globally by Chameides et al. [1987] to estimate the contribution of lightning to the NO\textsubscript{x} budget. More recently, Ridley et al. [1996] used observations of NO\textsubscript{x} in two New Mexico thunderstorms to extrapolate a global lightning production of NO\textsubscript{x} in the range of 3–5 Tg(N) yr\textsuperscript{-1}, and Huntrieser et al. [1998] found a most likely value of 4 Tg(N) yr\textsuperscript{-1} based on observations of thunderstorms during a field study over Germany and Switzerland. One shortcoming of cloud-scale field studies is that aircraft measurements can only be taken in the outer, more benign portions of the storm (typically in the anvil region), whereas much of the lightning activity occurs near the core of the storm. Another shortcoming of this approach is due to the large variability of individual thunderstorms; no individual thunderstorm can be representative of all thunderstorms occurring throughout the globe at all times.

For the present study a two-dimensional (2-D) cloud resolving model (the Goddard Cumulus Ensemble (GCE) model) was used to simulate production of NO\textsubscript{x} by lightning in a thunderstorm that occurred during the Stratosphere-Troposphere Experiment—Radiation, Aerosols, Ozone—Part A—Deep Convection (STERAO-A) field campaign. The primary objective of STERAO-A was to examine the effects of deep convection on the chemical composition of the middle and upper troposphere including the production of NO\textsubscript{x} by lightning and transport of NO\textsubscript{x} from the boundary layer. A complete description of STERAO-A is given by Dye et al. [2000]. The goals of our research were (1) to infer from the model simulations and measurements the relative production rates of NO\textsubscript{x} by IC and CG lightning for the July 12 storm during STERAO-A; (2) to assess the relative contributions in the storm anvil of convective transport of NO\textsubscript{x} from the boundary layer and NO\textsubscript{x} production by lightning; and (3) to simulate the effects of the lightning-generated NO\textsubscript{x} on subsequent photochemical ozone production. We have focused our efforts on a thunderstorm that occurred over southeastern Wyoming and northeastern Colorado on the afternoon and evening of July 12, 1996. This storm began as a series of isolated cells oriented in a west-to-east direction and propagated toward the east-southeast during the period from 2000 to 2300 UTC, with the westward most cell dominating by the end of this period. Lightning first occurred in the storm a few minutes after 2100 UTC. The storm took on a more three-dimensional (3-D) character after 2300 UTC and became ill-suited for simulation by a 2-D model. Therefore the three-hour period between 2000 and 2300 UTC is the focus of the numerical modeling efforts of this study.

The model includes passive transport (no chemical reactions) of NO\textsubscript{x}, CO, O\textsubscript{3}, and three hydrocarbons (ethane, propane, and propene), as well as a parameterized source of NO\textsubscript{x} from lightning based on observed CG and IC flash rates (determined using data from the National Lightning Detection Network™ (NLDN) and a VHF interferometer operated by the French Office Nationale d’Etudes et de Recherches Aérospatiales (ONERA)). Prior to and during this thunderstorm, aircraft measurements of temperature, dew point, wind, and mixing ratios of CO, NO, O\textsubscript{3}, and hydrocarbons were made by both the National Atmospheric and Oceanic Administration (NOAA) WP-3D and the University of North Dakota (UND) Cessna Citation II. The WP-3D also measured mixing ratios of alkyl nitrates, peroxyacetyl nitrate (PAN), peroxides, formaldehyde, and a variety of other species. The meteorological and chemical measurements in advance of the convection were used for model initialization in conjunction with rawinsonde and ozonesonde data. Model-generated winds were used to advect the trace gases. Distributions of CO and NO\textsubscript{x} from the model were then compared with observations of these species taken from the UND Cessna Citation II aircraft while flying in the anvil of the thunderstorm. We used the CO simulation to assess transport in the model. The NO\textsubscript{x} simulations were used to determine the fraction of the NO\textsubscript{x} in the cloud due to transport from the boundary layer versus that from production by lightning, as well as to estimate appropriate values of \( P_{CG} \) and \( P_{IC} \).

Section 2 of this paper describes our lightning NO\textsubscript{x} parameterization for the GCE model, and section 3 presents our simulation of lightning NO\textsubscript{x} generation and transport for the July 12 STERAO-A storm. Specific results are outlined in section 4, while section 5 discusses the results in further depth and demonstrates the importance of lightning NO\textsubscript{x} for downstream ozone production.

### 2. Lightning NO\textsubscript{x} Parameterization

Pickering et al. [1998] developed a lightning NO\textsubscript{x} parameterization for use with the 2-D GCE Model. The lightning NO\textsubscript{x} source parameterization described here is an improved version. Observed CG and IC flash rates are used rather than empirically derived flash rates. The vertical distribution of lightning NO\textsubscript{x} is recognized as being an important consideration for global CTMs [e.g., Ridley et al., 1996]. This is because the lifetime of NO\textsubscript{x} in the upper troposphere is much greater than in the lower troposphere and also because the ozone production efficiency per NO\textsubscript{x} molecule is much greater in the upper troposphere compared to the lower troposphere [Lu et al., 1987]. Pickering et al. [1998] showed that the bulk of lightning NO\textsubscript{x} is located in the upper troposphere at the end of a thunderstorm; therefore the effective vertical structure of the lightning NO\textsubscript{x} source in CTMs should reflect these results. Their algorithm assumed that individual flashes are uniformly distributed in the vertical within separate CG and IC lightning regions. The new algorithm described below assumes nonuniform vertical distributions of CG and IC lightning NO\textsubscript{x} source (see section 2.2 for discussion of why nonuniform distribution is chosen).

#### 2.1. Change in NO Mixing Ratio Due to Lightning

The general equation for change in NO mixing ratio at a given altitude due to a single lightning flash (either CG or IC) is derived as follows. If the number of moles of NO\textsubscript{x} produced per unit length of lightning channel (those regions of the flash where there is sufficient current flow and heating of the air to form NO) at altitude \( z \) is given as \( n_{NO}(z) \), and the fraction of the total lightning channel length \( L \) contained between altitude \( z \) and \( z + dz \) is given as \( f(z)dz \), then the number of moles of NO\textsubscript{x} produced between altitude \( z \) and \( z + dz \) is given as

\[
dN_{NO}(z) = Lf(z)n_{NO}(z)dz. \tag{1}
\]

The change in mixing ratio \( q_{NO} \) is found by dividing (1) by the number of moles of air into which the lightning NO\textsubscript{x} is mixed, found from the ideal gas equation of state \( V = N_{air}RT/p \), \( V \) is volume, \( N_{air} \) is the number of moles of air, \( R \) is the universal gas constant, \( T \) is absolute temperature, and \( p \) is
The volume is given by $\Delta x \Delta y dz$ (where $\Delta x$ and $\Delta y$ are the horizontal dimensions of the volume into which the lightning NO$_2$ is added), and so the general equation for change in mixing ratio at altitude $z$ due to a single lightning flash is

$$\Delta q_{\text{NO}}(z) = \frac{LRT(z)f(z)n_{\text{NO}}(z)}{p(z)\Delta x \Delta y}. \quad (2)$$

The production of NO per unit length of lightning channel is a function of pressure, and some laboratory measurements have suggested a linear relationship over the range of pressures encountered in the troposphere [Wang et al., 1998b]. For simplicity, we assume that $n_{\text{NO}}$ is directly proportional to pressure with proportionality constant $C$, which allows pressure to be eliminated from (2) to yield

$$\Delta q_{\text{NO}}(z) = \frac{LCRT(z)f(z)}{\Delta x \Delta y}. \quad (3)$$

Equation (3) tells us that the change in mixing ratio at a given altitude due to a single lightning flash depends on the total NO produced by the flash (accounted for by $L$ and $C$), the vertical distribution of the channel segments, $f(z)$, the horizontal area over which the NO is mixed ($\Delta x \Delta y$), and the temperature at level $z$. This temperature dependence arises because as temperature increases there are fewer moles of air in a given volume, so adding a fixed number of moles of NO to the volume will result in a higher NO mixing ratio.

To apply (3) to a model simulation, the total length of the lightning channel and the proportionality constant $C$ must be known, or estimates of the total number of moles of NO produced by a lightning flash ($N_{\text{total}}$) must be used. Choosing the latter approach, (1) is integrated from the surface to the maximum altitude reached by the lightning channel ($z_T$) to give

$$N_{\text{total}} = \int_{z_0}^{z_T} Lf(z)n_{\text{NO}}(z)\,dz = LC \int_{z_0}^{z_T} f(z)p(z)\,dz, \quad (4)$$

which can be solved for $LC$. Equations (3) and (4) are the basis for computing the change in $q_{\text{NO}}(z)$ due to a single lightning flash within the model. Since the bulk of NO$_2$ produced by lightning is in the form of NO [Wang et al., 1998b], we do not consider production of NO$_2$.

### 2.2. Vertical Distribution of Lightning Channel Segments

Lightning channel segments are not uniformly distributed in the vertical, and the distribution is highly variable from storm to storm or even within an individual storm as it evolves [MacGorman and Rust, 1998b]. Owing to the location of the July 12 STERAO-A storm relative to the interferometer array, the vertical distribution of lightning channel segments could not be determined; instead, a physically and observationally reasonable form for $f(z)$ had to be estimated. Numerous studies with systems that map lightning channel segments (from both CG and IC flashes) show that the segments tend to be distributed vertically in either one or two layers [e.g., MacGorman et al., 1981; Taylor et al., 1984; Ray et al., 1987; Maier et al., 1995]. MacGorman et al. [1981] hypothesized that this layering is directly related to the vertical dipole that describes the general charge structure of the thunderstorm, and showed that individual IC lightning channels often have significant horizontal structure in each of the two layers, with little vertical channeling between the two layers. In some cases a pronounced vertical channel does connect the two layers of horizontal structure [Shao and Krehbiel, 1996]. Cloud-to-ground flashes also have horizontal structure, but it is usually confined to the lower layer [MacGorman and Rust, 1998]. On the basis of these studies the vertical distribution of the CG flash segments was estimated as a Gaussian distribution, given as

$$f(z) = \frac{1}{\sqrt{2\pi}\sigma} \exp \left[ -\frac{(z - \mu)^2}{2\sigma^2} \right] \quad (5)$$

where $\mu$ is the altitude of the maximum negative charge density and $\sigma$ is chosen such that 99% of the lightning channel segments lie between the ground and the cloud top. For CG flashes, $\mu$ was chosen as the altitude of the $-15^\circ\text{C}$ isotherm, a reasonable proxy for where the maximum negative charge is located [Houze, 1993]. For IC flashes, two Gaussian distributions were superimposed to achieve a bimodal distribution. The lower mode was at the altitude of the $-15^\circ\text{C}$ isotherm, while the altitude of the top mode was varied between model runs to test the sensitivity of the model to the vertical distribution chosen. Figure 1 shows examples of the CG and IC vertical channel segment distributions used in the model.

### 2.3. Horizontal Area for Lightning NO Placement

MacGorman and Rust [1998b] point out that based on numerous studies mapping lightning channels with respect to radar reflectivity [e.g., Mazur and Rust, 1983; Mazur et al., 1986; Proctor, 1983; Ray et al., 1987; Shao and Krehbiel, 1996; Taylor et al., 1984] most lightning channels correspond to areas of reflectivity >20 dBZ, except those channels near the top of the cloud or in anvils (which are most likely IC flashes). On the basis of these findings the NO produced by CG flashes was confined to regions of reflectivity greater than 20 dBZ as calculated from the model hydrometeor field. Thus $\Delta x$ (distance along the model axis) for CG flashes varies with altitude and is the cumulative distance along the model’s horizontal axis where reflectivity is greater than 20 dBZ. The NO produced by IC flashes is confined within the “visible cloud” defined as that region where the total hydrometeor mixing ratio exceeds 0.01 g/kg, so $\Delta x$ for IC flashes is the cumulative distance along the model’s horizontal axis where the total hydrometeor mixing ratio exceeds 0.01 g/kg and also varies with altitude.

Since the model is 2-D, it gives no information about the
cross-axis dimension of the clouds. To determine $\Delta y$ (distance across the model axis), radar images were analyzed to find a representative value for the width of the observed convection during the period of 2000–2300 UTC. On the basis of this analysis a value of 20 km was chosen.

2.4. Flash Rate Determination and NO Production per Flash

The CG flash rate was determined using data from the NLDN. The IC flash rate was then computed by subtracting the CG flash rate from the total flash rate determined from the VHF interferometer operated by ONERA (only flashes whose duration was greater than 100 ms were counted). The NO from lightning flashes was introduced into the model at intervals of 3 min. Figure 2 shows the time series of CG and IC lightning flash rates for the period from 2100 to 2300 UTC, which recorded 155 CG flashes and 1108 IC flashes. This figure also shows a time series of the IC/CG ratio, which was less than 10 over most of this 2-hour period (with the exception of three strokes of a CG flash and during the leader phase of an IC flash). Production of significant amounts of NO is thought to occur during the return strokes of a CG flash and during the leader phase of an IC flash [Price et al., 1997]. Production of NO in the corona phase of a lightning flash is thought to be negligible [Coppens et al., 1998]. Our understanding of NO production processes in lightning is, however, far from complete.

We have chosen to use two recent methods of estimating $P_{\text{CG}}$; one based on energy dissipation as related to peak current, and one based solely on peak current. The first estimate was based on Price et al. [1997], who give a formula for calculating the energy ($E_{\text{CG}}$ in joules) per CG flash from the peak current ($I_{\text{pe}}$ in A) as

$$E_{\text{CG}} = 1.823 \times 10^4 I_{\text{pe}}.$$  

(7)

The peak currents of all CG strokes during the thunderstorm were reported by the NLDN and had a mean of 15 kA (half that of the U.S. mean of near 30 kA [Wacker and Orville, 1999]), giving a mean energy of $2.7 \times 10^3$ J. Using a production of NO of the order of $10^{17}$ molecules NO J$^{-1}$ [Price et al., 1997], a value of $P_{\text{CG}} = 460$ mol flash$^{-1}$ is obtained.

The second method involved extrapolating the laboratory results of Wang et al. [1998b] to the observed lightning data. On the basis of their formula, at a peak current of 19 kA, the amount of NO produced per meter of lightning channel ($n_{\text{NO}}$) can be written as

$$n_{\text{NO}}(p) = a + bp$$  

(8)

where $a = 0.34 \times 10^{21}$ molecules m$^{-1}$ and $b = 1.30 \times 10^{16}$ molecules m$^{-1}$ Pa$^{-1}$. Using an empirical formula for pressure as a function of altitude [Lide, 1994, chap. 14], (8) can be written in terms of height as

$$n_{\text{NO}}(z) = a + c \left( \frac{z - A}{B} \right)^n$$  

(9)

where $z$ is in meters, $c = 1.30 \times 10^{16}$ molecules m$^{-1}$, $A = 4.43 \times 10^4$ m, $B = -4.95 \times 10^3$ m, and $a = 5.26$. If the total flash length, $L$, and vertical distribution of the lightning flash segments, $f(z)$, are known, then the number of molecules of NO produced per flash can be calculated by

$$P_{\text{CG}} = L \int_{z_0}^{z_T} f(z) n_{\text{NO}}(z) \, dz,$$  

(10)

where $z_0$ is the surface elevation and $z_T$ is the height of the top of the electrically active region of the storm. Combining (5), (9), and (10) yields a final expression for NO per CG flash of

$$P_{\text{CG}} = \frac{L}{\sqrt{2\pi} \sigma} \left[ a + c \left( \frac{z - A}{B} \right)^n \right] \exp \left[ -\frac{(z - \mu)^2}{2\sigma^2} \right] \, dz.$$  

(11)

For use in (11) we assumed values of $z_0 = 2$ km, $z_T = 12$ km. There is no consensus in the literature as to the “mean length” of a lightning channel; therefore we chose $L = 18$ km based.
on the distance of 5 km between the negative charge region and the ground, and a tortuosity of 3.6 [Wang et al., 1998b]. Multiplicity was assumed to be 3.5 [Thompson et al., 1984], and each subsequent return stroke was assumed to have one half the peak current of the first return stroke [Berger et al., 1975]. Numerical integration of (11) yields \( P_{CG} = 56 \text{ mol flash}^{-1} \). Scaling this value to the observed 15 kA peak current (using the formula for \( n_{IC}(I_o) \) from Wang et al. [1998b]) yields a value of 39 mol flash\(^{-1}\), an order of magnitude less than that obtained by using the method of Price et al. [1997].

The NO production from IC flashes suffers from even greater uncertainty than does that for CG flashes, owing mainly to lack of measurements of the peak current of IC flashes, as well as to an incomplete understanding of the mechanisms of NO\(_x\) production within an IC flash. This uncertainty is reflected in the many different values for the ratio of \( P_{IC}/P_{CG} \) seen in the literature (see section 1). For this study we assume \( P_{IC}/P_{CG} \) values of 0.1 to 1.0.

3. Synoptic Environment and Cloud Model Simulations

3.1. Storm Environment and Evolution

An east-west oriented cold front advanced southeast across the Wyoming-Colorado border and central Nebraska at 0900 UTC July 12, 1996, during the STERAO-A field campaign. Postfrontal cold air advection over the next 6 hours was directed across the panhandle of Nebraska, western Kansas, and along the Front Range of Colorado. Surface dew points over southern Wyoming and eastern Colorado indicated the existence of a strong moisture gradient which developed as a consequence of the front. By 0000 UTC July 13, easterly (up-slope) flow in the vicinity of the frontal boundary enhanced low-level pooling of moist air across northeastern Colorado and along the Front Range.

A short wave trough propagated southeast through southern Wyoming into northeast Colorado between 1200 UTC July 12 and 0000 UTC July 13. Cyclonic vorticity advection ahead of the short wave, along with midlevel cooling, enhanced the upward vertical velocity during this period. The low-level flow from the east-southeast at 2–5 m s\(^{-1}\) existed beneath a midlevel flow from the west-northwest at 7–10 m s\(^{-1}\), providing a favorable shear environment for the development of strong storms. A relatively low convective available potential energy (CAPE) of 710 J kg\(^{-1}\) m\(^{-2}\) was present in the observed sounding (shown in Figure 3). The bulk Richardson number was approximately 7, consistent with a low CAPE environment with some shear.

Initial afternoon convection developed along the Front Range in the vicinity of the Cheyenne Ridge in southeastern Wyoming. Elevated heating of the surface in this region, and pooling of low-level moisture associated with the trailing portion of the frontal boundary, served to focus the first towering cumulus in the vicinity of the ridge. Subsequent convection organized along outflow boundaries produced by the initial convection, and propagated to the east southeast. During the early stages of growth the convection was composed of a series of multicell storms which exhibited little organization and weak anvil growth. The organization became better defined between about 2140 UTC and 2300 UTC as the convection acquired a distinct linear structure oriented in a west to east line (Figure 4). This period of linear organization as the convection moved away from the higher terrain is the focus of our 2-D numerical simulation efforts.

3.2. Goddard Cumulus Ensemble (GCE) Model

A complete description of the GCE model is given by Tao and Simpson [1993]. It has been used extensively for convective storm-scale research and studies of convective transport of trace gases in the tropics and midlatitudes [e.g., Tao et al., 1991, 1993, 1996; Pickering et al., 1990, 1992, 1998; Scala et al., 1990; Stenchikov et al., 1996]. The version of the model used in the present study is described as follows. The vertical grid consists of 31 points with a maximum resolution of 220 m in the lowest levels, decreasing to near 1 km near the top of the domain at approximately 20 km. The 2-D horizontal domain consists of 514 grid points, with the central 430 having a fine resolution of 1 km. The outer grid points are horizontally stretched, and open lateral boundary conditions are used.

The version of the model used at the University of Maryland was recently modified to include nonhomogeneous initial conditions using forecast fields from the National Centers for Environmental Prediction (NCEP)Eta model, interpolating the Eta model fields to the GCE model grid. A “no slip” bottom boundary condition is used, with no surface fluxes of heat or moisture; the terrain simply acts as an aerodynamic barrier over which the air must flow. The modified version of the model also includes transport of four chemical species run at the same 3 s time step as the other processes in the model. The model does not include any reactions between the chemical species, but does include a source term for generation of NO\(_x\) by lightning (see section 2). The lightning-generated NO\(_x\) is treated as a separate species from the NO\(_x\) initially present (which is passively transported without sources or sinks) so that comparison can be made between the NO\(_x\) fields with and without lightning at any grid point at any time step in the model.

Figure 3. Composite environmental sounding of temperature, dew point, and wind (m s\(^{-1}\)), plotted in skew T versus log p format, using data from Fort Morgan, Colorado, at 1956 UTC and aircraft data taken by the NOAA WP-3D in undisturbed air ahead of the storm at about 2200 UTC.
3.3. Initial Conditions

The meteorological variables in the model were initialized using fields from the NCEP Eta model 12-hour forecast valid at 0000 UTC on July 13, 1996. These fields were chosen over the 6-hour forecast valid at 1800 UTC as the former were more representative of the actual conditions observed at 1800 UTC. An Eta 0-hour analysis at 1800 UTC would have been preferred, but was unavailable from the NCEP archives. The model was oriented along a west-to-east cross section corresponding with the Colorado-Wyoming border (41°N latitude).

Owing to an observed moisture deficiency of over 3 g/kg (water vapor mixing ratio) in the Eta model boundary layer (similar deficiency documented by Dunn and Horel [1994] and since corrected by NCEP after STERAO-A), the horizontally averaged vertical moisture profile was modified using the observed moisture profiles taken from the composite sounding (described in section 3.1). The modification brought the moisture field in line with observations while preserving horizontal gradients.

Both the NOAA WP-3D and UND Cessna Citation II carried instruments for measuring CO, O₃, and NO. Attempts to measure NO₂ from both aircraft were also made, but the data were determined to be unreliable. Instead, values of NO₂ were estimated based on the photostationary state assumption [Leighton, 1961]. This assumes that the production of NO₂ through reaction of NO with O₃

\[ \text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2 \]  \hspace{1cm} (12)

is nearly instantaneously balanced by its destruction through photolysis

\[ \text{NO}_2 + hv \rightarrow \text{NO} + \text{O}. \]  \hspace{1cm} (13)

This assumption is good as long as the reactions of peroxy radicals with NO can be ignored, which is generally the case in the free troposphere and in the boundary layer away from large sources of organic radicals. By setting the time derivative of the NO₂ concentration equal to zero an equation for the concentration of NO₂ can be derived in terms of the NO and O₃ concentration, which is

\[ [\text{NO}_2] = \frac{k_{12}[\text{NO}][\text{O}_3]}{J_{13}}, \]  \hspace{1cm} (14)

where \( k_{12} \) is the rate coefficient for (12) and \( J_{13} \) is the photolysis rate for NO₂ in (13). The NOAA Aeronomy Laboratory provided the estimates of NO₂ from the WP-3D measurements of NO and O₃ using measured values of radiative fluxes to calculate the NO₂ photolysis rates. Estimates of NO₂ from the UND Cessna Citation II aircraft measurements of NO and O₃ in regions free of cloud were calculated at the University of Maryland. For these calculations, values for \( k_{12} \) based on DeMore et al. [1997] were used, and \( J_{13} \) was calculated using actinic fluxes computed with the radiative transfer algorithm described by Stamnes et al. [1988], based on the solar zenith angle and column ozone amount.

The aircraft data showed significant horizontal variability in the concentrations of CO, O₃, hydrocarbons, and NOₓ. We chose to initialize the model using profiles of these species taken from a spiraling ascent of the WP-3D aircraft in undisturbed air about 100 km ahead of the storm at about 2200 UTC. In these initial profiles the boundary layer was cleaner in

---

**Figure 4.** Visible satellite imagery centered over northeastern Colorado for 2215 UTC July 12, 1996. North is upward on figure. Labels included for geographic reference are CO, Colorado; WY, Wyoming; NE, Nebraska; KS, Kansas; B, Boulder; C, CHILL radar site; FM, Fort Morgan. The convection of interest is located just north of the Colorado-Wyoming border and is oriented in a west-to-east direction.
terms of CO and NO\(_x\) than in other profiles taken by the aircraft on this flight; however, these cleaner profiles are the only ones that we were certain were uninfluenced by convective outflow. The remainder of the O\(_3\) profile came from an ozone-sonde launched at Boulder, Colorado, at 1635 UTC. A few data points at higher altitude in undisturbed air for NO\(_x\) were available from the UND Cessna Citation II aircraft. The remainder of the NO\(_x\) profile (in the upper troposphere and lower stratosphere) was estimated by scaling against the observed O\(_3\) profile because of the dominance of the stratospheric source of NO\(_x\) at these altitudes. The upper portion of the CO profile was constructed from measurements made over the northern Great Plains during previous summer field campaigns [e.g., Luke et al., 1992; Poulida et al., 1996].

3.4. Thunderstorm Simulation

Thunderstorm simulations are normally initialized with a single sounding, and convection is triggered by imposing a “warm bubble” or “cool pool” during the initial time steps. Usually, the model sounding must contain large amounts of CAPE in order to sustain convection [Emanuel, 1994]. We found that with our simulation, using nonhomogeneous initial conditions and terrain, convection was produced and sustained without a warm bubble or cool pool. Convective cells were aided in their development by the upslope flow. The convection was sustained, despite the relatively low CAPE, due in part to the horizontal gradients in the fields.

The thunderstorm simulated by the cloud model has to compare favorably with the observed thunderstorm because the transport and the lightning parameterization are dependent on storm structure. Even though the simulation generated sustained convection without imposing a warm bubble, in order to achieve a better comparison of the model convection with the observed storm (in terms of cloud top height), we chose to impose a weak warm bubble during the first 10 min of the simulation. The warm bubble was placed in the region where convection initiated in the simulation without the warm bubble. The warm bubble extended from longitudes 105°W to 106.25°W in the horizontal and from the surface to 2.75 km in the vertical. The heating rate in the warm bubble was 0.25°C min\(^{-1}\), resulting in a temperature perturbation of less than 2.5°C. After cessation of the warm bubble, the convection was allowed to develop on its own.

The simulated convection resembled the observed convection in several important respects. The maximum altitude of the 20 dBZ radar reflectivity contour derived from the simulation (Figure 5a) was within 0.5 km of the 14.5 km observed from the Colorado State University CHILL radar facility (Figure 6). The maximum model-derived reflectivity of 70 dBZ also compared favorably to the 65 dBZ maximum observation from the CHILL radar. Another factor for comparison is the difference in altitude between the maximum cloud top height and the average height of the anvil top. In the simulated storm this difference is roughly 3 km (see Figure 5a), while in the observed storm this depth is closer to 4 km (Figure 6). The time evolution of the simulated and observed convection was more difficult to compare since detailed radar analyses of the early portion of the storm were not available. The data that were available showed the evolution of the simulation to be similar to that of the observed storm. In both cases, 40 to 50 min after reaching maximum height the altitude of the 20 dBZ contour decreased to under 12 km. Figure 5b shows the model-derived radar reflectivity at the end of the simulation. An estimate of area-averaged rainfall rate of 15 mm h\(^{-1}\) was available from the CHILL radar at 2247 UTC. The area-averaged rainfall rates from the model simulation at 160 and 170 min (corresponding to ~2240 and 2250 UTC) were 16 mm h\(^{-1}\) and 11 mm h\(^{-1}\), respectively, comparing very favorably with the radar estimate.

Cell motion in the simulation was nearly twice as fast as the observed speed of 8 m s\(^{-1}\), the result of an overacceleration of the mean winds aloft in the model. Part of this overaccelera-

![Figure 5](image-url)
tion is likely due to incompletely specified initial conditions and the use of a 2-D approximation when it is likely that some 3-D forcing was present. The open lateral boundary conditions used in the GCE (and most other cloud and mesoscale models) may also be a factor. These boundary conditions are somewhat artificially formulated to minimize the reflection of outward propagating gravity waves, and even small errors in the horizontal velocity and pressure perturbation on the lateral boundaries can sometimes result in significant acceleration of the domain-averaged velocity [Klemp and Wilhelmson, 1978; Pielke, 1984].

As mentioned in section 1, the observed convection later reintensified and took on a 3-D character. Not surprisingly, the 2-D cloud model did not simulate this reintensification since it could not adequately simulate the 3-D motion fields nor account for the time evolution of the larger-scale dynamical forcing (e.g., moisture convergence) that was responsible for the intensification of the observed storm. Thus our study concentrates only on the first 3 hours of this event (2000–2300 UTC).

To facilitate comparison of the simulated lightning NO\textsubscript{x} distribution with that observed, the time that lightning was first observed in the storm is used as a reference mark. This occurred within 5 min of 2100 UTC according to the observations from the NLDN and the ONERA interferometer. We use the concept of Price and Rind [1992], who gave an empirical relation relating flash rate and vertical velocity, to determine when lightning should first occur in the model simulation. Lightning is assumed not to occur in the model until vertical velocities exceed 7 m s\textsuperscript{-1} (vertical velocities lower than this give extremely low flash rates in the empirical formula), which occurred about 60 min into the model run.

4. Results

4.1. Transport of CO

CO is a good tracer for transport in thunderstorms due to its approximately 1 month tropospheric lifetime [Dickerson et al., 1987]. We used CO as a passive tracer in the GCE model to verify how well the simulation handled transport from the boundary layer into the anvil of the storm. Figure 7 compares the initial CO vertical profile with the CO field after 180 min of simulation. The simulated convection has transported boundary layer values of CO into the anvil of the storm. To compare this with observations, data were obtained from a spiraling ascent through the anvil of the storm by the UND Cessna Citation II shortly after 2300 UTC, which is approximated by the end time (180 min) of the model simulation. We are certain that the aircraft was measuring within the anvil of the storm based on radar observations and the notes from the aircraft crew; however, it is not clear exactly where in the anvil of the simulated convection a vertical profile should be taken for comparison with the aircraft observations. Instead of picking a specific location in the model domain for comparison with the aircraft observation, we use horizontally averaged profiles taken from the model in the anvil of the simulated convection. The domain of the averaging extended 89 km (1° of longitude at this latitude) upwind from the downwind edge of the anvil cloud as determined by the model hydrometeor field (all further model profiles discussed are horizontally averaged using this method, unless stated otherwise). Therefore each level in the profile represents an average of 89 horizontal grid cells. The variability of the model profile is indicated by calculating the square root of the variance at each model level. Figure 8 shows the comparison of the model profile of CO mixing ratio compared with the aircraft observations. The model profile is located near the center of the aircraft observations (though it is lower than observations in the 10 to 11 km layer) and shows that CO in the anvil is enhanced by over 20 ppbv due to convective transport. This result demonstrates the model's ability to represent transport of a chemical species from the boundary layer to the upper troposphere within the thunderstorm. It also verifies that our 2-D frame of reference provides a good approximation of the 3-D flow structure which must have existed in the observed storm.

Figure 6. Radar reflectivity factor (dBZ) from the Colorado State University CHILL radar facility at Greeley, Colorado, at 2234 UTC July 12, 1996, presented in range-height indicator (RHI) format. This was near the time of maximum intensity during the early phase of the convection. The maximum reflectivity in this panel is 65 dBZ. Axis orientation is nearly east-west, showing two cells along the convective line.
4.2. NO\textsubscript{x} Field Without Lightning Source

Figure 9 compares the initial profile of NO\textsubscript{x} with that after 180 min of simulation with no lightning source of NO\textsubscript{x}. As opposed to the initial CO profile, which was well mixed in the boundary layer to over 3 km mean sea level (msl), the initial NO\textsubscript{x} profile exhibited a very sharp vertical gradient. Both CO and NO\textsubscript{x} have dominant surface sources, and both are subject to rapid mixing in the boundary layer. The difference in their profiles is due to their differing lifetimes. CO has a lifetime measured in months, while in a photochemically active boundary layer the lifetime of NO\textsubscript{x} is measured in hours [Ridley et al., 1996]. The NO\textsubscript{x} lifetime is of the same order as the convective mixing timescale, which is given as $\tau = z/w_*$ [Stull, 1988], where $z$ is the depth of the boundary layer and $w_*$ $\sim 1$ m s$^{-1}$ is the vertical mixing velocity. This means that loss of NO\textsubscript{x} is occurring as it is being mixed upward, resulting in a vertical gradient in the boundary layer. The simulation did not transport very much NO\textsubscript{x} from the boundary layer into the anvil because this sharp gradient resulted in a relatively thin layer of enhanced NO\textsubscript{x} near the surface (discussed further in section 5.2). The simulation also suggested that there was some slight downward transport of stratospheric NO\textsubscript{x} into the upper part of the thunderstorm.

To compare the model-simulated NO\textsubscript{x} with observations measured from the Cessna Citation II in the anvil of the storm, the NO measurements must be converted into a value of NO\textsubscript{x} by estimating the amount of NO\textsubscript{2} present, again using (14). This is complicated by the fact that, though the photolysis rate for NO\textsubscript{2} can be readily calculated under clear-sky conditions, within the anvil the photolysis rate can either be increased or decreased depending on the optical depth of the cloud and the solar zenith angle [Kelley et al., 1995]. On the basis of Madronich [1987] we estimate that the ratio $J_{(13)\text{anvil}}/J_{(13)\text{clear sky}}$ will be somewhere in the range of 0.5 to 4.0. Figure 10a shows the range of the mixing ratio of NO\textsubscript{x} versus altitude based on the Cessna Citation II NO and O\textsubscript{3} data (both were averaged to 10 s) assuming that the in-cloud enhancement of $J_{(13)}$ is between 0.5 and 4.0. This figure also shows the model profile of NO\textsubscript{x}, which has considerably less NO\textsubscript{x} than the observations. One possible source of enhanced NO\textsubscript{x} in the anvil would be downward transport of stratospheric air; however, this would be expected to be accompanied by elevated values of ozone, which were not present in the aircraft observations. Therefore we conclude that the difference between the model and observations is due to the lack of a source of NO\textsubscript{x} in the model from lightning.
4.3. NOx Field With a Lightning Source

Several model simulations were run using various values of NO production per flash. The first simulation used $P_{\text{CG}} = 460$ mol flash$^{-1}$ (based on Price et al. [1997]; also see section 2.4) and $P_{\text{IC}} = 46$ mol flash$^{-1}$, in line with the assumption of Price et al. [1997] that $P_{\text{IC}}/P_{\text{CG}} = 0.1$. The IC flashes were assumed to have a bimodal distribution with the upper mode corresponding to the altitude where ambient temperature is $-40^\circ$C. The model run with the lightning source has a weak NO$_x$ plume blowing downwind in the anvil of the storm. Figure 10b compares the model profile of NO$_x$ with that derived from the Cessna Citation II data. The inclusion of the lightning source of NO$_x$ gives better agreement with observations of NO$_x$ in the anvil than does transport alone. The model plume is broad compared to the observed plume (likely due to the nearly 1 km vertical resolution in the model at anvil altitudes, as well as the horizontal averaging). The greatest difference between the model and observations is in the amplitude of the NO$_x$ plume, with the model falling short of the aircraft measurements. This cannot be attributed to the use of an averaged profile from the model, since the maximum NO$_x$ mixing ratio in the anvil of the simulated storm was still less than 500 parts per trillion by volume (pptv).

The model was next run using a $P_{\text{CG}} = 460$ mol flash$^{-1}$ as before, but with $P_{\text{IC}} = 230$ mol flash$^{-1}$ ($P_{\text{IC}}/P_{\text{CG}} = 0.5$). The results from this simulation compare very favorably with those from the observations (Figure 10c) in terms of the magnitude and altitude of the NO$_x$ plume in the anvil. The NO$_x$ field after 180 min for this run (Figure 11) shows the pronounced local maxima in the anvil of the storm. The amount of NO$_x$ contained in a column of air in the anvil (between altitudes of 7.4 to 11.2 km) was computed from the observations and from the model results (see Table 1). The observations indicate that the column NO$_x$ is in the range of 350–510 $\mu$g(N) m$^{-2}$. The model has a range of 420–820 $\mu$g(N) m$^{-2}$, indicating considerable overlap with the range computed from the observations. The model well simulates the observed mixing ratios in the upper portion of the anvil, but overestimates the NO$_x$ mixing ratio in the 7.5–9.5 km layer.

These results suggest that for this particular storm the IC flashes were significant producers of NO and that the production of NO from an IC flash was of the order of 200 mol flash$^{-1}$. This result is consistent with Stith et al. [1999], who estimated an NO production of between 20 to 200 mol flash$^{-1}$ by analyzing a combination of aircraft NO measurements and VHF interferometer data for the July 9 and 10 STERAO-A storms. At the altitudes at which their measurements were taken, it is probable that the majority of their data are from IC flashes, especially when considering that the IC/CG ratio was 3 during the period of the July 9 storm that they sampled, and for the portion of the July 10 storm all flashes were IC. Their measurements likely underestimate the production of NO per flash since some of the NO produced by a lightning flash will rapidly convert to NO$_2$ via reaction with ozone. With this in mind, the $P_{\text{IC}}$ value from our numerical experiments (230 mol flash$^{-1}$) is consistent with the upper end of the range of their calculations.

Our results show that $P_{\text{IC}}$ is likely of the order of 200 mol flash$^{-1}$, but we have not yet analyzed what effect changes in our assumption for the production by CG flashes has on the results. To this end, the model was run with $P_{\text{IC}} = 230$ mol flash$^{-1}$ and $P_{\text{CG}} = 2300$ mol flash$^{-1}$ ($P_{\text{IC}}/P_{\text{CG}} = 0.1$). These results (Figure 10d) show that although the amplitude of the NO$_x$ plume is of the right order, the NO$_x$ in the lower anvil is severely overestimated, and the peak is at a lower altitude than observed. The column amount of NO$_x$ in the anvil from the model is 550–1300 $\mu$g(N) m$^{-2}$, a range that is much higher than the observations indicate. We also ran the model with $P_{\text{CG}} = P_{\text{IC}} = 230$ mol flash$^{-1}$ (i.e., $P_{\text{IC}}/P_{\text{CG}} = 1$; Figure 10c). The results are very similar to the run using $P_{\text{CG}} = 460$ mol

![Figure 9](image-url)  
(a) Initial profile of NO$_x$ in model; (b) model NO$_x$ field (pptv) after 180 min of simulation with no lightning source.
Figure 10. Model profiles of NO$_x$ through the anvil (solid line) for the various model runs after 180 min compared with the range of NO$_x$ mixing ratios derived from measurements from the Cessna Citation II aircraft in the anvil (shaded area). Brackets indicate the square root of the variance of model values used for horizontal average. The upper mode of the IC flash distribution was at -40°C except for Figure 10f. Model runs are (a) transport only, no lightning, (b) $P_{CG} = 460, P_{IC} = 46$; (c) $P_{CG} = 460, P_{IC} = 230$; (d) $P_{CG} = 2300, P_{IC} = 230$; (e) $P_{CG} = P_{IC} = 230$; (f) same as Figure 10c only with upper mode of IC flash distribution at -30°C. Units for $P_{CG}$ and $P_{IC}$ are mol flash$^{-1}$.

The column amount of anvil NO$_x$ in this case ranges from 405 to 760 μg(N) m$^{-2}$, giving slightly more overlap with the range of anvil NO$_x$ calculated from observations; however, due to the uncertainties discussed in section 5.1, we cannot argue that this simulation is significantly better than the one using $P_{CG} = 460$ mol flash$^{-1}$ and $P_{IC} = 230$ mol flash$^{-1}$, and for this reason we continue to use the latter in our further discussion regarding vertical dis-
distribution of lightning NO$_x$ mass, comparison of NO$_x$ transport versus production by lightning, and effects of lightning NO$_x$ on photochemical ozone production.

We can impose some constraints on $P_{CG}$ based on these results. The peak current of IC flashes is estimated to be an order of magnitude less than for CG flashes (though current measurements aloft are scarce, Burkett et al. [1988] measured peak currents in the range of 1–3 kA), suggesting that the total channel length of an IC flash would need to be more than 10 times that of a CG flash for $P_{IC}$ to exceed $P_{CG}$. In the literature, assumptions for the ratio of $P_{IC}/P_{CG}$ vary widely (see section 1), but lie between zero and unity. Of course, this does not rule out the possibility of $P_{IC}$ exceeding $P_{CG}$; however, from a conservative standpoint, by constraining $P_{IC}/P_{CG}$ to be less than or equal to 1 a lower limit of $P_{CG}$ of the order of 200 mol flash$^{-1}$ is obtained. An upper limit for $P_{CG}$ of the order of 500 mol flash$^{-1}$ can be justified by the model results shown in Figure 10 and in the lightning NO$_x$ mass values presented in Table 1 because values greater than this significantly overestimate the NO$_x$ in the anvil.

One final set of experiments were performed using a value of $P_{CG} = 39$ mol flash$^{-1}$ based on the laboratory measurements of Wang et al. [1998b] (see section 2.4) and a value of $P_{IC}/P_{CG} = 1$. In these simulations the NO$_x$ production by the model was almost negligible compared with the previous results. NO$_x$ mixing ratios did not exceed 400 pptv anywhere within the simulated cloud, and in the anvil the NO$_x$ plume exhibited a mixing ratio of less than 300 pptv. The column amount of anvil NO$_x$ from this simulation was only 140–210 µg(N) m$^{-2}$, significantly lower than that calculated from the observations. We thus conclude that $P_{CG}$ must have been of the order of 200 mol flash$^{-1}$ or greater.

5. Discussion

5.1. Uncertainties

The results of this study indicate that production of NO$_x$ by IC flashes was the dominant factor in determining the distribution of NO$_x$ in the anvil of the thunderstorm studied. The results also suggest that the IC flashes had a NO$_x$ production of the order of 200 mol flash$^{-1}$ and that production by CG flashes was of the order of 200 mol flash$^{-1}$ $< P_{CG} < 500$ mol flash$^{-1}$. Probably the largest source of uncertainty is due to the sensitivity of the lightning NO$_x$ mixing ratios to the horizontal area over which the lightning NO$_x$ is assumed to be instantaneously mixed. The change in mixing ratio at a given altitude due to a lightning flash is inversely proportional to this horizontal area (see equation (3)). The model also assumes that mixing occurs instantaneously over this area, instead of diffusing more slowly. Therefore local peaks in NO$_x$ concentration resulting from individual flashes are not represented in the model; instead we are simulating the ensemble effects on NO$_x$ of over 1200 flashes that occurred during the period of interest.

The unknown vertical distribution of the lightning channel segments is another source of uncertainty. The results in section 4.3 are based on the assumption that the IC channel segments are distributed bimodally, with modes at the altitudes corresponding to temperatures of $-15^\circ$C and $-40^\circ$C. To test the sensitivity of the results to the vertical distribution of channel segments, a run was made similar to the second simulation discussed in section 4.3 ($P_{CG} = 460$ mol flash$^{-1}$, $P_{IC}/P_{CG} = 0.5$), only lowering the upper mode of the IC channel segment distribution to an altitude corresponding to a temperature of $-30^\circ$C. These results, compared in Figure 10f, indicate the simulations are sensitive to the vertical distribution of lightning channel segments. By lowering the upper mode of the flash distribution the maximum in the NO$_x$ plume occurs at a lower altitude. The actual vertical distribution of channel segments in this storm is unknown, but the results suggest that it was similar

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Anvil Column NO$_x$, µg(N) m$^{-2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Observations</td>
<td>350–510*</td>
</tr>
<tr>
<td>Model: transport only (no lightning)</td>
<td>80–95</td>
</tr>
<tr>
<td>Model: $P_{CG} = 460$ mol flash$^{-1}$; $P_{IC} = 46$ mol flash$^{-1}$</td>
<td>180–330</td>
</tr>
<tr>
<td>Model: $P_{CG} = 460$ mol flash$^{-1}$; $P_{IC} = 230$ mol flash$^{-1}$</td>
<td>420–820</td>
</tr>
<tr>
<td>Model: $P_{CG} = 2300$ mol flash$^{-1}$; $P_{IC} = 230$ mol flash$^{-1}$</td>
<td>550–1300</td>
</tr>
<tr>
<td>Model: $P_{CG} = 230$ mol flash$^{-1}$; $P_{IC} = 230$ mol flash$^{-1}$</td>
<td>405–760</td>
</tr>
<tr>
<td>Model: $P_{CG} = 39$ mol flash$^{-1}$; $P_{IC} = 39$ mol flash$^{-1}$</td>
<td>140–210</td>
</tr>
</tbody>
</table>

*For observations the range represents the minimum and maximum amounts estimated from observations of NO and photostationary steady state calculations for NO$_x$. For model simulations the range represents the mass from horizontally averaged NO$_x$ profile plus or minus the square root of the variance of the mass from the individual profiles comprising the average profile.
Figure 12. Percent of NO\textsubscript{x} in simulation that is the result of lightning after 180 min ($P_{\text{CG}} = 460$ mol flash$^{-1}$, $P_{\text{IC}} = 230$ mol flash$^{-1}$).

to that used in the initial model runs (modes at $-15^\circ C$ and $-40^\circ C$) since the altitude of the NO\textsubscript{x} plume in the anvil from this run agreed best with the observations.

There is uncertainty in the initial vertical profile of NO\textsubscript{x} used in the model since the measured distribution was not horizontally uniform; however, we believe that the profile we used is the most representative and has the least chance of having been contaminated by the storm. An assessment of this uncertainty is achieved by rerunning the model using an initial NO\textsubscript{x} profile constructed by averaging all the measurements taken by the WP-3D aircraft data at each altitude. This averaged profile contained about 60% more NO\textsubscript{x} in the boundary layer than did the single profile used for the model runs discussed in sections 4.2 and 4.3. Use of the profile with larger boundary layer values of NO\textsubscript{x} increased NO\textsubscript{x} in the anvil of the storm due to transport by up to 60 pptv, an increase of about 30% if only NO\textsubscript{x} transported from the boundary layer is considered. This increase becomes less than 10% when both transport and lightning generation of NO\textsubscript{x} are considered, indicating that the uncertainty from this source is small.

Using a 2-D model to simulate an inherently 3-D process such as the development of a thunderstorm and subsequent production and transport of NO\textsubscript{x} also introduces some uncertainty. Not only does the 2-D model assume a fixed width of the storm with altitude, but it also assumes that the width of the storm does not change with time. Additionally, 3-D advective and diffusion effects are not treated. We anticipate simulating this and other STERAO-A events with the 3-D version of the GCE model in the near future to reduce this uncertainty.

5.2. Lightning Production of NO\textsubscript{x} Versus Transport From Boundary Layer

Figure 12 shows the percentage of NO\textsubscript{x} in the model fields after 180 min that is attributed to lightning ($P_{\text{CG}} = 460$ mol flash$^{-1}$, $P_{\text{IC}} = P_{\text{CG}} = 0.5$). Over 80% of the NO\textsubscript{x} in the anvil of the storm is due to lightning, while less than 20% is attributed to transport of NO\textsubscript{x} from the boundary layer. The percentage maximizes at over 90% in the midtropospheric part of the cloud where the CG mode and lower IC mode coincide. Down drafts in the model cause percentages in the lowest kilometer to reach 50% at the rear of the storm and a small amount of lightning NO\textsubscript{x} to reach the surface.

The relative lack of boundary layer NO\textsubscript{x} transported to the anvil of the storm is due to the sharp vertical gradient and very shallow layer of NO\textsubscript{x} initially present near the surface. Backward trajectories from the maxima of CO and NO\textsubscript{x} mixing ratios at the end of the model run were computed and indicated that these air parcels originated in the 2–4 km (msl) region, well above the surface elevation (which is under 2 km).

The initial layer of high CO mixing ratios was much deeper than that for NO\textsubscript{x}, explaining why boundary layer values of CO were transported into the upper troposphere (see Figure 7), but values of NO\textsubscript{x} in the upper troposphere due to transport alone are much smaller than the boundary layer values (see Figure 9).

The separation of the upper tropospheric CO and NO\textsubscript{x} maxima in the model simulation (compare Figures 7b and 11) illustrates the difference in the processes by which these maxima result (convective transport from the boundary layer versus middle and upper tropospheric production by lightning). If transport were dominant over the lightning production of NO\textsubscript{x}, then the maxima of CO and NO\textsubscript{x}, whose concentrations are both initially highest in the boundary layer, would be expected to be nearly coincident.

5.3. Vertical Distribution of Lightning NO\textsubscript{x} Mass

Ridley et al. [1996] point out that determining the vertical distribution of the lightning NO\textsubscript{x} mass is just as important as determining the amount of NO\textsubscript{x} produced. This is due to the large increase in the lifetime of NO\textsubscript{x} with altitude in the troposphere (in the upper troposphere NO\textsubscript{x} can persist for a week or more).

Figure 13 shows the distribution of NO\textsubscript{x} mass from the model run for the July 12 STERAO-A storm using the current version of the lightning algorithm and $P_{\text{IC}}/P_{\text{CG}} = 0.5$. It does not have the “C” shape of the vertical profiles (maxima near ground and in anvil) from Pickering et al. [1998]. One explanation for this is that unlike the storm cases from that previous work, this simulated storm contained only a very weak rear...
downdraft to transport the lightning NO\textsubscript{x} downward. Also, by using a nonuniform vertical lightning channel distribution, less of the lightning NO\textsubscript{x} was initially placed near the ground. Another factor may be that the simulations of the convective cases presented by Pickering et al. [1998] were carried through the dissipation stage of the storms, allowing more time for NO\textsubscript{x} mass redistribution than in the present study. It is inappropriate to draw any further general conclusions from these differences based solely on this single storm simulation because there is a large amount of variability of individual thunderstorms from any sort of global mean, and the relative frequency of strong downdraft versus weak downdraft continental storms is unknown.

5.4. Effects on Downstream O\textsubscript{3} Production

Within the cloud, measurements from the Cessna Citation II showed no evidence of rapid production of ozone by electrical discharges or photochemistry or rapid loss due to heterogeneous chemical processes. However, upper tropospheric NO\textsubscript{x} from thunderstorm anvil outflow has the potential to catalyze the production of O\textsubscript{3} downstream. To assess the impact of the lightning NO\textsubscript{x} from this thunderstorm on subsequent ozone production, a one-dimensional (column) chemical model using the SMVGEAR II chemical solver [Jacobson, 1995] and a simplified tropospheric chemistry mechanism was initialized with anvil profiles of O\textsubscript{3}, NO\textsubscript{x}, and three hydrocarbon species (ethane, ethene, and propane) from the GCE model run with lightning ($P_{\text{CG}} = 460 \text{ mol flash}^{-1}$, $P_{\text{IC}} = 230 \text{ mol flash}^{-1}$) and without lightning. It was assumed that the anvil cloud immediately dissipated at the end of the GCE model run so that clear-sky conditions could be assumed, and the NO\textsubscript{x} was partitioned into NO and NO\textsubscript{2} using the photostationary state assumption (see section 3.3). In addition, the effects of anvil shear and turbulence in diluting the plume of lightning NO\textsubscript{x} are not included in this simulation, so it represents what is likely to be the maximum effect of the lightning NO\textsubscript{x} generated in the early phase of the July 12 STERAO-A storm on downstream ozone production.

Figure 14a shows a time-altitude (pressure) plot of the difference in ozone concentration between the simulation with lightning ($P_{\text{CG}} = 460 \text{ mol flash}^{-1}$, $P_{\text{IC}}/P_{\text{CG}} = 0.5$) and without lightning. Contours are in ppbv. (b) Time series of ozone at 277 hPa for these two simulations. Top curve is with lightning; bottom curve is without lightning.

Figure 14. (a) Time-pressure cross section of the difference in the ozone concentration between the simulation with lightning ($P_{\text{CG}} = 460 \text{ mol flash}^{-1}$, $P_{\text{IC}} = 230 \text{ mol flash}^{-1}$) and without lightning. Contours are in ppbv. (b) Time series of ozone at 277 hPa for these two simulations. Top curve is with lightning; bottom curve is without lightning.
production rates from as low as 1 ppbv $\text{d}^{-1}$ to as high as 28 ppbv $\text{d}^{-1}$ under a variety of storm types, locations, and conditions.

6. Summary and Conclusions

We have shown that the ensemble of 1108 IC lightning flashes was the dominant producer of NO in the thunderstorm observed on July 12, 1996, during the STERAO-A field campaign. By assuming that $P_{\text{IC}} = 230$ mol flash$^{-1}$ the model was able to realistically simulate the observed NO$_x$ distribution in the upper portion of the anvil of the storm. The results allow us to estimate values for production of NO$_x$ by a CG flash of the order of $200 < P_{\text{CG}} < 590$ mol flash$^{-1}$ for the July 12 storm, which had a mean peak current for CG flashes of 15 kA. Therefore NO$_x$ production per CG flash in this storm appears to be of the order of magnitude estimated using the method of Price et al. [1997] and an order of magnitude greater than Wang et al. [1998b]. Values of the ratio $P_{\text{IC}}/P_{\text{CG}}$ in the range 0.5–1.0 yielded the best comparisons with NO$_x$ observations. Individual thunderstorm variability in terms of lightning flash rate, IC to CG flash ratio, peak current, lightning channel length, and a host of other parameters preclude an extrapolation of these results to estimate the global annual total production of NO$_x$ by lightning. Detailed chemical and lightning observations from more storms are needed to reduce these uncertainties.

The NO$_x$ in the anvil of the thunderstorm appeared to be dominated by lightning production relative to transport from the boundary layer. Only a small quantity of boundary layer NO$_x$ reached the anvil because the storm’s updraft drew air primarily from the upper part of the boundary layer where NO$_x$ mixing ratios were initially quite small. The upper tropospheric maxima of CO and NO$_x$ in the simulation were separated horizontally and vertically, illustrating the differences in the processes which led to their formation (convective transport versus lightning production). Up to 7 ppbv $\text{d}^{-1}$ greater net photochemical ozone production was computed in the upper tropospheric outflow of this storm when NO$_x$ from lightning was considered, indicating the importance of gaining more knowledge concerning the nature of the lightning source of NO$_x$.

Acknowledgments. This research was supported under National Science Foundation (NSF) grants ATM 9627179 (KEP) and ATM 9628702 (JRS), a University of Maryland Graduate Fellowship, and a NASA Earth System Science Graduate Fellowship. We thank Wei-Kuo Tao of NASA/GSFC for providing the GCE model and for helpful advice on its modification and application. Data collection by the UND Cessna Citation II was supported under NSF grant ATM 9634125 (JLS). We thank Tom Ryerson, Paul Goldan, and David Parrish of the NOAA Aeronomy Laboratory for providing chemical measurements from the NOAA WP-3D aircraft, Karsten Baumann of NCAR (now at the Georgia Institute of Technology) for the CO data taken from the UND Cessna Citation II, and Sam Oltmans of the NOAA Climate Monitoring and Diagnostics Laboratory for providing ozoneonde data from Boulder, Colorado. We express our appreciation to Adrian Tuck (NOAA Aeronomy Laboratory) and Steve Rutledge (Colorado State University), who with one of the authors (JD) organized and coordinated the STERAO-A field project. We also thank Steve Rutledge for providing the NLDN and CHILL Radar Facility data.

References


Madronich, S., Photodissociation in the atmosphere, 1. Actinic flux


A. DeCaria and K. Pickering (corresponding author), Department of Meteorology, University of Maryland, 3433 Computer and Space Sciences Bldg., College Park, MD 20742. (pickerin@atmos.umd.edu)

J. Dye, B. Ridley, and J. Stith, National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO 80307.

P. Laroche, Office Nationale d’Etudes et de Recherches Aérospatiales, 92322 Chatillon Cedex, France.

P. Laroche, Office Nationale d’Etudes et de Recherches Aérospatiales, 92322 Chatillon Cedex, France.

J. Scala, The Weather Channel, 300 Interstate North Parkway, Atlanta, GA 30339.

G. Stenchikov, Department of Environmental Sciences, Rutgers—State University of New Jersey, 14 College Farm Rd., New Brunswick, NJ 08901-8551.

(Received July 27, 1999; revised January 4, 2000; accepted January 10, 2000.)