NO signatures from lightning flashes

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Abstract. In situ measurements of cloud properties, NOx, and other trace gases were made in active thunderstorms by two research aircraft. Concurrent measurements from a three-dimensional (3-D) VHF interferometer and the 2-D National Lightning Detection Network were used to determine lightning frequency and location. The CHILL Doppler radar and the NOAA-WP-3D Orion X band Doppler radar were also used to measure storm characteristics. Two case studies from the (STERAO) Stratosphere-Troposphere Experiments: Radiation, Aerosols, and Ozone project in northeastern Colorado during the summer of 1996 are presented. Narrow spikes (0.11–0.96 km across), containing up to 19 ppbv of NOx, were observed in the storms. Most were located in or downwind of electrically active regions where the NO produced by lightning would be expected. However, it was difficult to correlate individual flashes with NO spikes. A simple model of the plume of NO from lightning is used to estimate NO production from the mean mixing ratio measured in these spikes. The estimates range from $2.0 \times 10^{20}$ to $1.0 \times 10^{22}$ molecules of NO per meter of flash length.

1. Introduction

Lightning is thought to be a major natural source of NOx ($\text{NO + NO}_2$), an essential ingredient in the production of ozone in the troposphere. However, estimating the amount of NOx produced by lightning is a difficult proposition because of the many variables that influence the amount produced [e.g., Price et al., 1997, and references therein]. In addition, direct in situ measurements of NOx from lightning are relatively rare, especially for those measurements that can be directly related to lightning characteristics, such as flash type, flash duration, and flash rate. These types of measurements are needed because the NOx production is expected to be a function of the type of lightning flash and altitude. For example, an intracloud (IC) flash is thought to produce less NOx than a cloud-to-ground (CG) flash because of the lower air densities and lower energies per flash [e.g., Goldenbaum and Dickerson, 1993; Wang et al., 1998]. On the other hand, IC flashes are generally more frequent than CG flashes. Gallardo and Cooray [1996] suggest that cloud-to-cloud (which includes IC) flashes may be as effective as CG flashes at producing NOx. Calculations of global NOx production by lightning often utilize a global flash rate [e.g., Ridley et al., 1996, and references therein], but there is relatively limited geographical information on the vertical distribution of the energy from these flashes.

The location where NOx is formed and transported is also of great importance because NOx has a longer lifetime in the upper troposphere than at lower altitudes. NOx formed in the updraft of thunderstorms is likely to be dispersed into the upper troposphere or lower stratosphere, while that formed in the lower troposphere (or transported there by downdrafts) is more quickly removed from the atmosphere.

The development of remote sensing techniques for measuring lightning flashes, such as the National Lightning Detection Network (NLDN), satellite optical sensors, radio interferometry, and time of arrival techniques, allow individual flashes to be located and some of the properties of individual flashes to be determined. For example, the NLDN can determine the peak current and polarity (negative or positive) of CG strokes. In principal, lightning interferometers and time of arrival systems are able to distinguish CG from IC strokes and provide some measure of the velocity of the flash, which can assist in identifying flash type.

In this paper we report on the locations and characteristics of NO plumes which we attribute to NO produced from individual lightning flashes. We describe where they were located in the storms and contrast them with NO mixing ratios in other regions of the storms. The NLDN and a lightning interferometer made concurrent measurements of the flashes. We examine the lightning measurements to see how they correlate with the locations of the NO plumes. Finally, we use a simple model of the lightning channel to estimate the production of NO per meter of flash length.

2. Instrumentation and Description of the Experiment

Measurements were made in summertime thunderstorms during the 1996 Stratosphere-Troposphere Experiments: Radiation, Aerosols, and Ozone (STERAO) Deep Convection project in northeastern Colorado (J. E. Dye et al., An overview of the STERAO–Deep Convection Experiment with results for the July 10 storm, submitted to Journal of Geophysical Research, 1999; hereinafter referred to as submitted paper). The major instrumentation systems for this program include the National Oceanic and Atmospheric Administration (NOAA)
WP-3D research aircraft, the University of North Dakota (UND) Citation research aircraft, the Colorado State University CHILL Doppler radar, and the French Office National d’Études et de Recherches Aérospatiales (ONERA) three-dimensional (3-D) VHF interferometer [Laroche et al., 1994]. Below we briefly describe the major instruments used in this study and the results from some major case studies.

The UND Citation carries a set of standard aircraft instrumentation for measurement of state parameters (a Rosemount model 102 temperature probe, a Rosemount 1201FI pressure transducer, and a chilled mirror dewpoint instrument) and a five-hole radome gust probe/Inertial Navigation System for measuring 3-D wind and turbulence. Three particle measuring systems (PMS) probes measured cloud particle size distributions at a 4 Hz sampling rate. Cloud particle concentrations reported in this paper are from the PMS 2DC, 1DC, and 1DP probes. They represent the total concentration of particles greater than the minimum detectable size, which is in the range of 30–50 μm for the C probes, depending on particle type (particles <100 μm are poorly sampled, however). These data represent the ice particle concentrations in the cloud because ice was the predominant cloud particle type at the cold temperatures where sampling was conducted. The concentration of ice particles is a good indicator of the presence of cloud because ice was abundant in the regions that were sampled. The Citation sampled mainly in the upper troposphere, and the WP-3D sampled the lower levels up to about 5 km.

NO was measured with a fast-response chemiluminescence instrument similar to that described by Ridley and Grahek [1990]. A Scintrex LOZ3 Eosin Y chemiluminescent analyzer was used for ozone measurements. The uncertainty in the NO and ozone measurements is estimated to be 12 pptv ± 5% and 10%, respectively. The NO instrument was calibrated by in-flight dynamic dilution from a standard cylinder, and the ozone analyzer was calibrated on the ground using a reference ozone generator (Dasibi model 1008-PC).

The NOAA WP-3D aircraft carried several custom instruments for measurements of NO and ozone. These included an instrument for NO which was also based on the design by Ridley and Grahek [1990] and calibrated by in-flight dynamic dilution from a standard cylinder. A fast-response chemiluminescence ozone instrument, based on the reaction of ozone with excess NO, was used for ozone measurements and compared against a standard UV absorption unit. The uncertainty for these NO and ozone instruments is estimated to be 30 pptv + 15% and 0.03 ppbv + 3%, respectively. NO2 mixing ratios out of cloud were calculated from the photostationary–steady state relationship, using measured NO mixing ratios and the photodissociation rate constant for NO2 calculated from dual radiometer-measured UV irradiances using the methods described by Madronich [1987] and Davis et al. [1993].

The Colorado State University CHILL Doppler radar (located at Greeley, Colorado) is an 11 cm multiparameter radar with a beam width of 1.1° and a pulse length of 150 m. However, the main source of Doppler data for STERAO was the X band radar carried on the NOAA WP-3D. For this paper, we will use the reflectivity data from CHILL to describe the storm characteristics. Future studies of the STERAO cases will use the WP-3D radar data to derive storm airflow and the microphysical state of the hydrometeors.

The ONERA interferometer consists of two stations, one that detects the azimuth of VHF sources and a second that detects both azimuth and elevation. This allows the 3-D determination of VHF radio signals emitted by different parts of lightning discharges to be determined. Because positive discharges along the lightning channels do not emit strong VHF radiation, the interferometer detects primarily negative leader and recoil streamer processes. Thus not all parts of a lightning flash are measured. Information on the system is found in the work by Laroche et al. [1994] and Mazur et al. [1997], while Lang [1997] (available at http://olympic.atmos.colostate.edu/~lang/) provides a summary of the performance of the system during STERAO. The vertical resolution is nominally 0.5°. However at elevation angles less than 8° and greater than 42°, measurements were seriously compromised. As discussed by J. E. Dye et al. (submitted paper, 1999), storms at 50 km range provide reliable VHF source information for altitudes greater than 8.5 km, but this altitude is higher for more distant storms. Because most storms were at a greater range and because of the lack of adequate vertical resolution for most cases, we will use only the horizontal data in this paper. For STERAO a horizontal resolution of about 2 km was obtained within the main sampling lobes, with better resolution for closer storms. However, this resolution degraded for weak signals or for areas near the baseline and at the upper and lower parts of the lobes (J. E. Dye et al., submitted paper, 1999). The operational area of the interferometer extended from just north of the Wyoming and Nebraska border, and south as far as Greeley, Colorado, and east from the foothills of the Rocky Mountains about 150 km. This placed it in the northeastern coverage area of the CHILL radar.

The interferometer detects VHF source emissions along the lightning channels of both CG and IC flashes. These signals arise from negative leaders, K changes (recoil streamers), or spider discharges [see Laroche et al., 1994; Mazur et al., 1997]. It has difficulty detecting signals from return strokes or positive breakdown. Because return strokes are thought to produce a significant portion of the NO from a lightning flash, this is a significant limitation. Software developed by the ONERA research group and the National Center for Atmospheric Research (NCAR) is used herein to identify flashes. The software identifies time and position of individual VHF sources and uses the time, distance, and calculated velocity between sources to identify different flashes. A procedure to distinguish CG from IC was also implemented, based upon altitude; however, the degraded vertical resolution at low altitudes caused Lang [1997] to abandon this technique and compute the IC flash rate by taking the difference between the ONERA flash rate and the NLDN flash rate.

In this paper we show the locations of individual VHF sources as received by the interferometer. Typically, a large number of VHF sources are associated with a single flash, and the distribution of the sources in space gives an approximate 3-D map of the lightning flash, although not all portions of the flash produce an equal number of VHF sources. However, only horizontal maps could be constructed for this study, as discussed above. For comparison, we also plot some concurrent data from the NLDN, which provides one ground strike location for each CG flash detected. The performance of the NLDN is described by Cummins et al. [1998]. For the STERAO project it should detect 90% of CG flashes, with a median location accuracy of 500 m.
3. Case Studies

3.1. July 9 Case

3.1.1. Citation measurements. On July 9, 1996, there was widespread upslope cloudiness over eastern Colorado. Around 2130 (all times are universal time coordinated, UTC) a moderate thunderstorm with a large stratiform extent formed over the northern foothills of the Colorado Front Range and then moved south-southeast into the operation area along a line roughly parallel to the mountains. The mean wind during the sampling at midaltitudes (6 –7 km; all altitudes are given in altitude above mean sea level) was from 300\° at 12 m s\(^{-1}\). The CHILL radar reflectivity and 18 min of the Citation track are given in Figure 1. From 2245 to 2315, before the aircraft sampling, the storm had two cells with maximum radar reflectivities of 55–60 dBZ near the melting level, and it was slightly more organized. These two cells produced the anvil that was sampled later by the Citation. The horizontal radar return during the period of aircraft sampling indicated a widespread stratiform region containing patches of weak embedded convection. Reflectivities at 4.5 km were up to 45 dBZ (Figure 1a). The sampling was done near the tops of these weak cells (Figure 1b) and in the stratiform remains of the earlier storm that drifted southeast over the plains. This area was able to sustain embedded convection.

The Citation penetrated the storm initially at 2328 at a temperature of \(-16\) to \(-17\)°C, at 7 km altitude, while heading to the northern side of the storm (pass 1). A climb was commenced at 2337, and a turn toward the downwind anvil region was started at 2342, after reaching the northern side of the storm. Between 2348 and 0034, passes 2–5 were made through the anvil between 7.9 and 10.5 km altitude (Figure 2).

At 2330, just after entering cloud, a flash was observed in the cloud ahead of the aircraft and recorded in the flight notes. A few minutes later, several narrow plumes of NO (about 100 –750 m across) were encountered (Figure 3), including one containing a maximum of nearly 19 ppbv.

The small NO spikes on the south end of pass 1 correlate with the location of a small cell, located 22 km east of CHILL (Figure 1b), which was sampled near its top. The highest NO spike was also located near the northern lobe of that cell. The last set of spikes at 2335:30 was north of the cell and just east of higher reflectivity.

Data from this and the later passes are given in Figure 4. The largest spikes of NO were located in the cloud region that contained the highest concentration of both small and large particles observed in this storm. The region also had small amounts of liquid water. The liquid water concentrations (Figure 3) were near the detection limit of the instrument (which...
also produces a slight response when struck by ice particles. However, examination of the ice particle images from the PMS 2DC probe indicated that, in the region that contained spikes, the ice particle images exhibited a mixture of single particles, aggregates, and rounded particles characteristic of small graupel or heavily rimed particles (Figure 4b). Thus the data show an abundance of ice particles, the presence of graupel, and small amounts of supercooled liquid water. These are the microphysical ingredients thought to be necessary to produce charge separation [Dye et al., 1988]. There is recent evidence [Proctor, 1991] that both CG and IC lightning frequently originate in this temperature region. Thus the region where the highest NO mixing ratios were observed is a likely region for charge separation and lightning to be occurring. This was also an area where strong electrical activity was found by the lighting detection systems, particularly intracloud lighting as described below, which is consistent with lightning being the source of the NO spikes.

The spikes during pass 1 were much narrower with larger NO mixing ratios than those found in the anvil regions (Figure 4) during passes 2–5. This suggests that the aircraft was passing through or near the source of NO for pass 1. Pass 2, which was made in the lower anvil and stratiform region at 7.9 km, exhibited the lowest NO mixing ratios, while passes 4 and 5, which were near the top of the anvil at 9.1 and 10.3 km, exhibited the highest NO mixing ratios in the anvil, near 2 ppbv. These observations may be due to the lower temperatures (e.g., −40°C during passes 4 and 5) or possibly higher photolysis rates of NO₂ in the upper anvil, which would both produce a higher NO/NO₂ ratio. In addition, the results of Ridley et al. [1996] indicate preferential transport of NO₂ to the upper cloud regions of thunderstorms. Size sorting of the observed ice particles is also evident in Figure 4, as large particles were absent from the upper anvil at the farthest downwind range (pass 5).

### 3.1.2. WP-3D measurements.

The WP-3D sampled the region southeast of the storm between 2230 and 2330, at the lower levels between 2.5 and 5.0 km (Figures 5 and 6). The wind at 2.7 km was from the east at 7 m s⁻¹, so the lower portions of this region were likely to be flowing toward the storm. A major portion of the flight track was in cloud, so the cloud base was below 2.5 km in that area. No major sources of NOx were found (Figure 6). The plume from the WP-3D exhaust may have drifted into the region that was later sampled by the Citation between 2329 and 2338, but for the closest pass it would have had to traverse about 4 km vertically over a 30 min time period. It seems unlikely that it would have maintained a coherent enough plume structure to produce the sharp spikes that were observed from the Citation.
3.1.3. Interferometer and NLDN measurements. The storm was quite electrically active just before it was sampled, with 77 flashes detected by the ONERA interferometer and 16 by the NLDN during 2320–2336, the 16-min period preceding the last encounter with an NO spike during pass 1. The VHF sources from flashes that extended into the region of the NO spikes are presented in Figure 7, along with the locations of the spikes. During this time period, five of the 16 CG flashes recorded by the NLDN were near pass 1 and also detected by the interferometer (Figure 7). Many of the flashes in this storm extended over 40–50 km horizontally, nearly the entire width of the storm. Such large horizontal extents are typical of spider lightning, which is commonly observed in stratiform storm regions.

One flash, which was not detected by the NLDN, occurred at 2331:31, less than 3 min before the highest mixing ratio NO spike was observed. It was centered near the large spike but also had elements near the other spikes (small open boxes in Figure 7). Because it was not detected by the NLDN, it was likely to have been an IC flash.

We can conclude from the above that these NO spikes were located in a region of the cloud where active IC and CG flashes were present. The interferometer data also suggest that the horizontal extent of the flashes was quite broad, much more so than one might anticipate if only the data from the NLDN were available. Thus the interferometer data suggest that NO could have been produced over a wide area of the cloud by both IC and CG flashes, and portions of the NO plume from these flashes were likely observed. Because of the limited horizontal resolution and lack of vertical placement of VHF signals at this range from the interferometer baseline, it is difficult to correlate individual flashes with NO spikes, although the interferometer data suggest possible candidates.

3.2. July 10 Case Study

3.2.1. Citation measurements. On July 10, 1996, at 2100 an isolated thunderstorm developed near Kimball, Nebraska.

The storm was multicellular initially but became uncellular with characteristics of a low precipitation supercell at about 0100 (J. E. Dye et al., submitted paper, 1999). The Citation sampled the storm between 2245 and 0100, during a period of active development of the cellular features of the storm. The CHILL radar return from the storm at 2310:11–2314:07 is given in Figure 8, along with the partial track of the Citation. Two strong cells were evident, oriented in a northwest to southeast direction. The larger cell to the southeast was the main cell associated with the well-defined anvil that was sampled during the flight. The northwestern cell was lower during this time, but both cells had strong radar reflectivity and electrical activity. This storm had unusually high cloud tops near 16.5 km (Figure 8) and apparently penetrated well into the lower stratosphere. Sampling was done in the northern part of the anvil and the upshear anvil associated with the taller cell to the southeast from 2245 to 2303. From 2313 to 0020 a series of cross-anvil passes were made, between 12 and 8 km altitude, to obtain a cross section of the anvil about 50–60 km downwind from the main cell. Following these passes, a spiral up through the anvil near the center of the cross-anvil passes was conducted.

At least three distinct spikes of NO were sampled close to the active cells (Figure 9a). A small spike of NO was observed at 2249:54 very near the eastern edge of the anvil at 10.6 km. A second spike was found at 2259:47, as the aircraft began a
penetration in the upwind anvil of the largest cell, in the region between the two cells, which had filled in with cloud at the sampling altitude of 11.3 km. Following that pass a cross-anvil pass was made (Figure 8) at 11.5 km, just downwind of the large cell, that encountered a 4 ppbv mixing ratio spike of NO at 2307:44. A fourth spike was found at 2348:44 (Figure 9b) during one of the cross-anvil passes at 10.7 km. Given the typical measured wind speeds in the storm of \(20 \text{ m s}^{-1}\) from 290°, the exhaust from the Citation should have drifted downwind of subsequent passes during most of the sampling.

High ozone mixing ratios, characteristic of lower stratospheric air, were found on the southern side of the storm (Figure 9). The large vertical extent of this storm may have contributed to bringing down stratospheric air. Somewhat similar transport of stratospheric air by vigorous convective storms has been reported by Poulida et al. [1996]. Except for the occasional spikes, the maximum mixing ratios of NO found in the upper portion of the anvil were about 1 ppbv and somewhat less than the 2 ppbv found in the upper anvil of the July 9 case.

3.2.2. WP-3D measurements. While the Citation was sampling the storm, the WP-3D was doing step-up legs just southwest of the storm in the inflow region of the storm (Figure 10). Major sources of NO\(_x\) were not observed, and NO\(_x\) mixing ratios at the lowest levels were just over 1 ppbv (Figure 11). The cloud base below the storm was not measured; however, the lifting condensation level from a sounding made at 2000 from the Fort Morgan airport (about 75 km east of the CHILL radar) was at 2.8 km.

3.2.3. Interferometer and NLDN measurements. Data from the interferometer are presented in Figures 12 and 13.
along with the location of the NO spikes and the Citation. During the period before the spikes were observed, the storm produced almost exclusively IC lightning, with only a few CG strokes detected by the NLDN prior to 2350. IC lightning was abundant, with flash rates ranging from about 5 to 20 per minute between 2230 and 2350 (J. E. Dye et al., submitted paper, 1999). CG activity increased substantially after 2350 but was associated with the weaker cell to the northwest [Lang, 1997]. The first spike at 2249:54 was located downwind of the storm core and well away from the interferometer activity. The second spike, at 2259:47, was adjacent to the activity in the northern cell, which had substantial electrical activity at the time the spike was observed. The spike with the greatest NO, at 2307:44, was located just downwind of the electrical activity from the main cell, about 10–15 min downwind of the main activity. The fourth spike at 2348:44 (Figure 13) was in the anvil near two flashes that occurred at 2333:49 (crosses in Figure 13) and 2346:06 (small boxes in Figure 13). Interferometer signals from these two IC flashes were found in the main electrical region but also extending from the core into the northeast portion of the anvil.

### 4. NO Production by Lightning

A summary of the characteristics of the major NO spikes is given in Table 1. The peak, mean, and standard deviation of the NO mixing ratios for each spike are given (standard deviations for spikes only 1 s long are omitted). The means and standard deviations of the NO mixing ratios adjacent to the plume are given for 10 s of data (just over 1 km of flight track) adjacent to the plume (5 s on each side). In the last column of Table 1 the spikes are classified as A, B, or C, according to their proximity to known lightning flashes in time and space. Category A are those that are most likely to be associated with lightning flashes. In these cases, lightning was observed within a few minutes before the observed NO spike and at a location that would be likely to drift to where the NO was observed. Those in category B are spikes where lighting was observed at least 5 min prior to the spikes and at a location that would be likely to drift to within 5–10 km of where the spikes were observed. Category C refers to spikes that were well outside the interferometer lobes or far from observed interferometer sources. We cannot absolutely rule out the possibility that another source, such as another aircraft, might have produced some of these plumes; however, most aircraft would avoid these regions because of the proximity of the storms and lightning.

We seek to estimate the lightning production rate in terms of the molecules of NO produced per meter of flash length. To estimate this, the NO plume is modeled as follows. Consider a cylindrical slab of differential thickness $dL$ along the plume axis (Figure 14). The volume of the slab is $(\pi/4)D^2dL$, where $D$ is the diameter of the plume at the location of the cylindrical slab. The volume of lightning-produced NO in the slab, $V_{NO}$, is given by $V_{NO} = (\pi/4)D^2CdL$, where $C$ is the average volume mixing ratio of NO in the slab due to the lightning. The volume of NO per unit direction along the axis of the plume is $V_{NO}/dL = (\pi/4)D^2C$. Converting from cubic meters of NO

### Table 1. Summary of NO Production Deduced From Individual Spikes

<table>
<thead>
<tr>
<th>Date</th>
<th>Time of Peak (GMT)</th>
<th>NO Peak, ppbv</th>
<th>Pressure, Pa</th>
<th>Plume Mean NO, ppbv</th>
<th>Plume s.d.,*</th>
<th>Background Mean NO, ppbv</th>
<th>Background s.d., ppbv</th>
<th>Mean NO Above Background, ppbv</th>
<th>Plume Diameter, m</th>
<th>Temperature, °C</th>
<th>NO Produced, molecules m⁻¹</th>
<th>Spike Category (See Text)</th>
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<td>July 9</td>
<td>2332:55</td>
<td>1.70</td>
<td>40900</td>
<td>1.03</td>
<td>0.43</td>
<td>0.29</td>
<td>0.08</td>
<td>0.74</td>
<td>753</td>
<td>-17</td>
<td>3.8E + 21</td>
<td>A</td>
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<td></td>
<td>2333:07</td>
<td>1.40</td>
<td>40900</td>
<td>1.00</td>
<td>0.33</td>
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<td>0.05</td>
<td>0.66</td>
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<td>A</td>
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<td>1.80</td>
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<td>0.65</td>
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<td>A</td>
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<td>4.50</td>
<td>40900</td>
<td>4.47</td>
<td>0.95</td>
<td>0.16</td>
<td>0.28</td>
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<td>0.20</td>
<td>0.21</td>
<td>0.04</td>
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<tr>
<td></td>
<td>2307:44</td>
<td>4.20</td>
<td>20900</td>
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<td>0.02</td>
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<tr>
<td>Mean</td>
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<td>3.8E + 02</td>
<td></td>
<td>2.5E + 21</td>
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Read 3.8E + 21 as $3.8 \times 10^{21}$.  
*Standard deviations (s.d.) are omitted for plumes 1 s long.
Figure 14. Sketch of NO plume geometry for calculating the NO production.

\[ P_{\text{NO}} = \frac{PA_C \pi D^2}{4RT} \]

where \( P \), \( A_c \), \( R \), and \( T \) represent the pressure, Avogadro's number, the universal gas constant, and the temperature, respectively.

We assume that the NO from the lightning has mixed with background levels of NO by the time it is sampled. To exclude the background contribution, the mean mixing ratios of NO adjacent to the plume ("background mean NO" in Table 1) are subtracted from the measured mixing ratios in the plume ("plume mean NO" in Table 1) to give C ("mean NO above background" in Table 1). The diameter of the slab, \( D \), is approximated by the horizontal dimension along the flight track of the observed NO spikes, which is computed from the elapsed time in the plume times the true air speed. This equation was used to compute the NO production listed in Table 1.

For fresh plumes in stable air, the plume diameters are likely to be less than the resolution of the NO measurements (1 s), such as the plumes at 2333:39, 2334:42, and 2335:39 on July 9 (Table 1). Because the instrument recorded the mixing ratio over a 1 s interval, it provides an average mixing ratio similar to what would occur if the actual plume were dispersed over the distance represented by 1 s of flight path. However, the computed NO production values in Table 1 should be underestimates for plumes that were traversed off-center, and overestimates for plumes that were not perpendicular to the path of the aircraft. In addition, a significant fraction of the NO that was produced should have reacted with ozone to form NO2. Because this fraction was not measured, these estimates should be substantially lower than the actual NO production.

Recently, Wang et al. [1998] have simulated natural lightning in the laboratory, using arc discharges with a waveform and current (\( \approx 30 \) kA) similar to lower values found in natural lightning. They measured NO production rates as a function of pressure, current, and energy. They also compared their measurements with the results from a modified version of the numerical model described by Goldenbaum and Dickerson [1993] with generally good agreement. They measured NO production rates (at 1 atm pressure) between about \( 5 \times 10^{20} \) and \( 3 \times 10^{21} \) molecules m\(^{-2}\) for peak currents between 10 and 30 kA, respectively. NO production was found to linearly increase with pressure. These values are comparable to the lower values in Table 1 but lower than the average values when pressure is taken into account. However, the higher values in Table 1 (e.g., at 2334:46 on July 9) were in regions where multiple strokes or flashes were likely; this might account for part of the differences between Table 1 and the results of Wang et al. [1998].

Lawrence et al. [1995] summarize previous work on NO lightning production and present averages of the existing estimates of the NO production rate per flash, based on theoretical, laboratory, and field studies. These averages are 6.7, 7.0, and 92 (in \( 10^{25} \) NO molecules per flash), respectively, with corresponding standard deviations of 10, 6.6, and 122. Estimates included in their review range from 0.5 to 300 (\( 10^{25} \) NO molecules per flash). Our average value in Table 1 of \( 2.5 \times 10^{21} \) molecules m\(^{-1}\), assuming flash lengths between 5 and 50 km (including some of the different parts of the flash), would produce 1.25–12.5 in units of \( 10^{25} \) molecules per flash. Thus, for reasonable flash lengths, the average NO production rate in Table 1 is within the range of existing estimates.

5. Discussion and Conclusions

The measurements in section 3 suggest that the NO signatures from individual lightning flashes, or portions of these flashes, can be found in some locations in thunderstorms. These chemical signatures of the lightning flash seem to be correlated with the locations of the flash, as observed by the interferometer and NLDN; however, it was not possible to unambiguously associate a given measurement spike with a particular stroke.

Spikes of NO were found during periods when the storm on July 9 was largely stratiform. Spikes were also found during more active convection on July 10, but in the anvil, where the cloud was also stratiform with relatively low turbulence.

Storm regions with strong convection, such as on July 10, contained multiple flashes, even for relatively short time periods. The NO formed by these flashes in vigorous updrafts should mix through these regions within a few minutes, as has been observed in tracer releases of small plumes of sulfur hexafluoride into growing convective clouds [e.g., Stith et al., 1990, and references therein]. However, in regions with less turbulence, such as portions of the anvils, NO plumes would remain intact longer, improving their chances of being sampled before being diluted too much to be identified.

The majority of the NO spikes were found near or downwind of regions with strong activity measured by the interferometer. However, at least one spike was observed outside of the active areas, near the edges of the storms. This may be a result of the inability of the interferometer to identify certain parts of the flash, such as positive breakdown or perhaps another source, such as an aircraft, produced the spike.

In some cases, the data from the interferometer were able to suggest flashes that had produced the observed spikes of NO. It is unfortunate that the interferometer was not able to provide better vertical resolution, which would allow one to eliminate the signals that were well separated vertically from the observed NO spikes. This would have reduced the seemingly large horizontal dispersion in the interferometer signals. Better resolution in the vertical and horizontal interferometer data should be possible with more sophisticated interferometers, and this would greatly improve the opportunities for these types of comparisons. It should be possible to define the
3-D extent of the flash and a measure of the intensity for different regions of the flash. When combined with concurrent measurements of NO$_3^-$, this will provide a much clearer picture of NO production from lightning, and it will allow for more realistic extrapolations of measurements at one location to regional or global scales.

We estimate the NO production to be in the range of $2 \times 10^{20}$ to $1 \times 10^{22}$ molecules m$^{-1}$, and these estimates would be increased somewhat if the contribution from NO$_2$ were available. Our estimates suggest a somewhat higher level of NO production than indicated by the recent measurements of Wang et al. [1998] but are within the previous estimates of Lawrence et al. [1995]. However, given the uncertainty in these types of estimates, the comparisons are encouraging. With better resolution of lightning flashes, future studies such as this one should be able to better correlate NO production to lightning type and characteristics.

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