



Measurements of NO_X produced by rocket-triggered lightning

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[1] We present the first direct measurements of NO_X generated by specific lightning sources. In July 2005, three negative lightning flashes were triggered using the rocket-and-wire technique at the International Center for Lightning Research and Testing (ICLRT) at Camp Blanding, Florida. The NO_X produced by these three rocket-triggered flashes was measured. The NO_X production per unit charge was between 2 and $3 \cdot 10^{20}$ molecules per meter per coulomb. The data show that the NO_X production is primarily from long-duration, steady currents, as opposed to microsecond-scale impulsive return stroke currents. This observation implies that cloud discharges, which transfer, via a steady current of the order of 100 A, larger charges than ground discharges, but do not contain return strokes, are as efficient as (or more efficient than) cloud-to-ground discharges in producing NO_X . **Citation:** Rahman, M., V. Cooray, V. A. Rakov, M. A. Uman, P. Liyanage, B. A. DeCarlo, J. Jerauld, and R. C. Olsen III (2007), Measurements of NO_X produced by rocket-triggered lightning, *Geophys. Res. Lett.*, *34*, L03816, doi:10.1029/2006GL027956.

1. Introduction

[2] An assessment of the global distribution of nitrogen oxides, mainly NO and NO_2 , generally referred to as NO_X , is required for a description of tropospheric chemistry and in the evaluation of the global impact of the anthropogenic emissions of nitrogen oxides [Crutzen, 1970; Johnston, 1971]. Lightning is one of the primary natural (as opposed to man-made) sources of nitrogen oxides, and it may be the dominant source in the troposphere in the Equatorial and Tropical South Pacific [Gallardo and Rodhe, 1997]. An accurate quantification of the nitrogen oxides generated by thunderstorm processes is essential to further development of chemical models of the troposphere and in the evaluation of the impact of anthropogenic nitrogen oxide emissions in the terrestrial atmosphere.

[3] A direct measurement of the NO_X generated by a natural lightning flash is impractical because of the difficulty of confining a section of the lightning channel in a given volume so that the NO_X emitted by that section of

the lightning channel can be examined. This is why, researchers have employed indirect methods [Tuck, 1976; Noxon, 1976; Chameides et al., 1977; Drapcho et al., 1983; Franzblau and Popp, 1989; Stith et al., 1999; DeCaria et al., 2000; Cook et al., 2000; Huntrieser et al., 2002] to quantify the NO_X generated by lightning flashes. Due to a large number of uncertainties involved, the estimates of global NO_X production by lightning available in the literature vary by two orders of magnitude, from about 1 to over 20 Tg(N) per year [Rakov and Uman, 2003, p. 510]. Recent estimates typically range from 2 to 20 Tg(N) per year [Zhang et al., 2003].

[4] In this paper, we present the first direct measurements of NO_X generated by specific sources in lightning flashes, although the lightning in our experiments was artificially initiated (triggered) from natural thunderclouds using the rocket-and-wire technique [Rakov, 1999]. The NO_X produced by rocket-triggered lightning was measured by isolating a 3-cm section of the lightning channel within a discharge chamber and passing the air containing NO_X generated by that channel section to an NO_X analyzer.

2. Experiment

[5] The experiment was conducted at the International Center for Lightning Research and Testing (ICLRT) at Camp Blanding, Florida during summer 2005. Lightning was triggered using the rocket-and wire technique, as noted above, which involves the launching of a small rocket extending a thin grounded copper wire toward a charged cloud overhead, when the measured electric field at ground is sufficiently high (typically higher than about 4 kV/m in absolute value). At the time that the rocket reaches a height of about 200 to 300 m, the enhanced electric field near the rocket tip results in a positively charged leader that propagates upward toward the cloud. This upward positive leader (UPL) vaporizes the trailing wire, bridges the gap between the cloud and ground, and establishes a steady current, referred to as initial continuous current (ICC), with a typical duration of some hundreds of milliseconds that effectively transports negative charge (typically some tens of coulombs) from the cloud charge source to the triggering facility. The UPL and ICC constitute the initial stage (IS) of a "classical" triggered-lightning discharge. After cessation of the initial continuous current, one or more downward dart-leader/upward-return-stroke sequences may traverse the same path to the triggering facility. Strokes (leader/return stroke sequences) in triggered lightning are thought to be similar to subsequent strokes (that is, all strokes following the first) in natural lightning. Using rocket-triggered lightning to study the production of NO_X is advantageous for the following two main reasons: (1) the time and the location of lightning strikes are controlled; and

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(2) it is possible to confine a section of the lightning channel in a given volume, so that no information and assumptions concerning the wind speed, wind direction, and dispersion of NO_X from the lightning channel are necessary to infer the NO_X production by that channel section. Additionally, the signal-to-noise ratio is expected to be considerably better than in natural-lightning studies.

[6] The rocket-triggered lightning was allowed to create an arc between two electrodes, one of which was connected to the rocket launcher on which the lightning channel terminates, and the other was connected to a grounded test object (used for a different experiment). The electrodes were separated by 3 cm and effectively served to isolate a 3-cm section of the lightning channel. The electrodes were made of copper with tungsten coating and had a hemispherical (3-cm radius) shape. The electrodes, forming a horizontal gap, were placed inside a cylindrical polyethylene chamber that completely enclosed a volume of 0.77 m³. Atmospheric air entered the chamber continuously through a 2 m long and 4 mm inner diameter Teflon tube terminated in a needle valve. Air was evacuated from the chamber using a 70 m long Teflon tube with the same inner diameter, filtered, using a 5-micron particle filter, and passed through a calibrated NO_X Analyzer (model 9841B, Monitor Labs). The gas flow rate was 0.60–0.64 dm³/min. At this rate, if one assumes uniform NO_X concentration in the 770 dm³ chamber, and a peak NO_X concentration of, say, 24 ppmv (parts per million by volume), it would take about 11 days for the NO_X concentration in the chamber to decrease to the background level. Since that kind of time was not available for either of the two measurements, we had to extrapolate measured concentration curves to the background level. The analyzer measured the concentrations of NO and NO_X using a chemiluminescence method, with the lowest detectable concentrations being about 1 ppbv (parts per billion by volume). The uncertainty in the NO_X concentration measurements is estimated to be about 10%. The number of NO_X molecules, N_{NO_X}, was calculated by integrating the entire measured concentration curve, after the background level was subtracted, using the following equation:

$$N_{\text{NO}_X} = \left[\sum (C(t) - B) \Delta t \right] \cdot 10^{-6} \cdot F \cdot (N_A/V), \quad (1)$$

where $C(t)$ is the measured NO_X concentration in parts per million by volume (ppmv) as a function of time, B is the long-term average measured background concentration of NO_X, found to be 0.005 ppmv, Δt is the time interval between the individual concentration measurements in minutes ($\Delta t = 1$ min was used in this study), F is the sample air flow rate in dm³/min (0.62 dm³/min), N_A is the Avogadro constant in molecules/mol ($6.0221415 \cdot 10^{23}$ molecules/mol), and V is the ideal gas molar volume in dm³/mol at the measured temperature and pressure (24.94 dm³/mol at 31 °C and 101.4 Pa). The error due to the use of the ideal gas molar volume is included in the overall error (about 10%) given above for the NO_X measurements. During the experiments all equipment was powered by a generator through an uninterruptible power supply (UPS). The current through the gap was measured some tens of meters from the rocket launcher, at the test object used for another experiment. The current was sensed with a non-

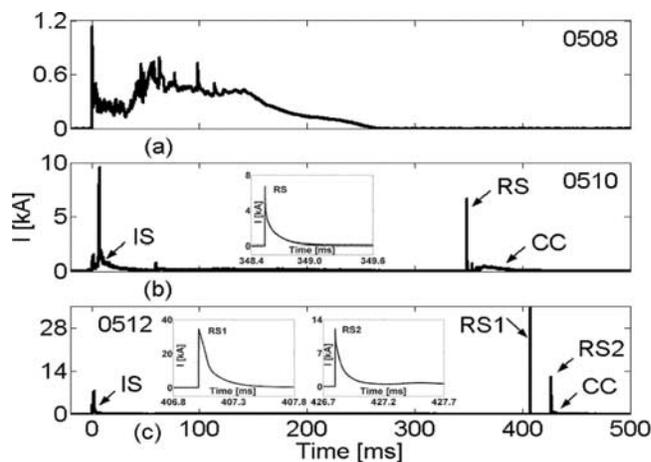


Figure 1. Current waveforms for (a) flash 0508 triggered on July 15, 2005, (b) flash 0510 triggered on July 31, 2005, (inset shows the RS current waveform on an expanded (1.2 ms) time scale), and (c) flash 0512 triggered on July 31, 2005, respectively (insets show RS current waveforms on expanded (1 ms) time scales).

inductive shunt whose output signals were transmitted via fiber-optic links to digitizers located (along with the NO_X Analyzer) in the launch control trailer.

[7] In this paper, we present measurements of NO_X produced by one flash containing only an initial-stage (IS) current (flash 0508 triggered on July 15, 2005) and by two flashes containing both IS and return strokes (flashes 0510 and 0512 triggered on July 31, 2005). Flashes 0510 and 0512 were triggered within eleven minutes of each other and contained one and two return strokes, respectively. The only return stroke in flash 0510 and second stroke in flash 0512 were followed by long (in excess of 40 ms) continuing currents. All three flashes lowered negative charge to ground.

3. Results

[8] Fortuitously, we were able to measure the number of NO_X molecules generated inside the discharge chamber by a steady IS current only (flash 0508) and also by a sequence of steady and impulsive current components (flashes 0510 and 0512). The overall current records for flashes 0508, 0510, and 0512 are shown in Figure 1. The return stroke current pulses are also shown on an expanded time scale (1.2 or 1 ms). The measured NO_X concentrations as a function of time are shown in Figure 2a for flash 0508 and in Figure 2b for flashes 0510 and 0512. In each case, the air from the discharge chamber was continuously sampled for about three days. Before integrating the entire curve in order to calculate the number of produced NO_X molecules using equation (1), the resultant concentration curves had to be extrapolated so that the concentration returns to the background level. In Figure 2a, the concentrations of NO and NO₂ are additionally shown. Concentration of NO₂ was found by subtracting measured concentration of NO from that of NO_X.

[9] The IS of flash 0508 had an average current of 289 A and a total duration of 266 ms. The largest current pulse of

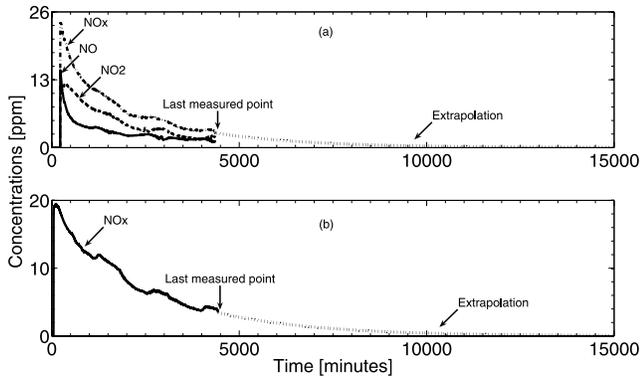


Figure 2. Measured concentrations of NO_x in ppmv as a function of time for (a) flash 0508, (concentrations of NO and NO_2 are additionally shown) and (b) flashes 0510 and 0512, respectively. Extrapolation from the last measured point, indicated by vertical arrow, to the background level, 5 ppbv, is shown by dashed line.

1.2 kA peak superimposed on the steady IS current is associated with the destruction of triggering wire [Rakov *et al.*, 2003; Olsen *et al.*, 2006], and the other superimposed pulses are of the M-component type [Rakov *et al.*, 2001]. This flash produced $6.1 \cdot 10^{20}$ NO_x molecules for the 3-cm channel length corresponding to $2.0 \cdot 10^{22}$ molecules per meter. The total charge transferred during the IS was 77 C. It follows that the NO_x yield for this IS was $2.6 \cdot 10^{20}$ molecules per coulomb per meter. Since an electric field of about 1 to 2 kV/m is needed to maintain an arc in air with a current of a few hundred amperes [King, 1961], the total energy dissipated in the 3 cm arc can be estimated to be about 2 to 4 kJ. Thus the NO_x yield of the IS in flash 0508 is about 1.5 to $3 \cdot 10^{17}$ molecules per joule, although this estimate should be viewed with caution, since we neglected the possible effect of metal vapor from the electrodes on the arc electric field. For comparison, the NO_x production by laboratory sparks has been estimated to be in the range from 0.2 to $4 \cdot 10^{17}$ molecules per joule [Rakov and Uman, 2003, p. 510].

[10] Flashes 0510 and 0512 occurred within about eleven minutes of each other, and therefore it was not possible to determine the NO_x production by each of these flashes separately. The IS of flash 0510 had an average current of 162 A, and its total duration was 266 ms. Interestingly the

largest IS current pulse superimposed on steady IS current had a peak of 9.6 kA, greater than the peak, 6.7 kA, of the following return stroke current pulse. The total charge transferred by this IS was 43 C. The total charge transferred by the only return stroke in this flash was about 0.5 C. This return stroke was followed by a continuing current which had a duration of about 87 ms and an average current of 138 A. The total charge transferred by this continuing current was about 12 C. The IS of flash 0512 had an average current of about 116 kA and a total duration of 361 ms. A current pulse of 7.6 kA peak was superimposed on the steady IS current. The total charge transferred by the IS current was about 42 C. The two return strokes in flash 0512 had peak currents of 34 and 12 kA, and the total charges transferred by them were 3.8 C and 0.8 C, respectively. The continuing current associated with the second stroke of flash 0512 had a duration of 59 ms and an average current of 100 A. This continuing current transferred about 5.9 C of charge. Flashes 0510 and 0512 together produced a total of $7.3 \cdot 10^{20}$ NO_x molecules in the chamber, which corresponds to $2.4 \cdot 10^{22}$ NO_x molecules per meter. Since the total charge transferred by flashes 0510 and 0512 was about 108 C, their total NO_x yield was about $2.2 \cdot 10^{20}$ molecules per coulomb per meter. This value is similar to the NO_x yield, $2.6 \cdot 10^{20}$ molecules per coulomb per meter, obtained for flash 0508, which contained only an IS and no return strokes. All results listed above are summarized in Table 1.

4. Discussion and Summary

[11] In the estimations of the global production of NO_x by lightning it is usually assumed that return strokes of ground flashes are the primary contributors, and contributions from cloud discharges are generally neglected [Chameides, 1986]. However, Gallardo and Cooray [1996] challenged this assumption (see also Rakov and Uman [2003, p. 509–511]). The results presented here indicate that relatively slow discharge processes, those occurring on time scales of milliseconds to hundreds of milliseconds, such as continuing currents in both cloud and cloud-to-ground flashes and other steady currents, can contribute significantly to the global NO_x production by lightning. Steady currents in rocket-triggered lightning are similar to those in natural cloud-to-ground lightning and in cloud discharges, with typical durations being in the tens to hundreds of milliseconds range and typical magnitudes of

Table 1. A Summary of Measurements of NO_x Produced by Three Rocket-Triggered Lightning Flashes^a

Flash ID	Lightning Process	Peak Current, kA	Duration, ms	Charge Transfer, C	Average Current, A	NO_x Molecules Per Meter	NO_x Molecules Per Coulomb Per Meter	NO_x Molecules Per Joule
0508 July 15, 2005	IS only	1.2	266.0	77	289	$2.0 \cdot 10^{22}$	$2.6 \cdot 10^{20}$ (Q = 77 C)	$1.5 - 3 \cdot 10^{17}$
0510 July 31, 2005	IS	9.6	266.0	43	162	$2.4 \cdot 10^{22}$	$2.2 \cdot 10^{20}$ (Q = 108 C)	-
	RS	6.7	1.0	0.5	-	$2.4 \cdot 10^{22}$	$2.2 \cdot 10^{20}$ (Q = 108 C)	-
	CC	-	87.0	12	138	$2.4 \cdot 10^{22}$	$2.2 \cdot 10^{20}$ (Q = 108 C)	-
0512 July 31, 2005	IS	7.6	361.0	42	116	$2.4 \cdot 10^{22}$	$2.2 \cdot 10^{20}$ (Q = 108 C)	-
	RS1	34.0	0.9	3.8	-	$2.4 \cdot 10^{22}$	$2.2 \cdot 10^{20}$ (Q = 108 C)	-
	RS2	12.0	0.4	0.8	-	$2.4 \cdot 10^{22}$	$2.2 \cdot 10^{20}$ (Q = 108 C)	-
	CC	-	59.0	5.9	100	$2.4 \cdot 10^{22}$	$2.2 \cdot 10^{20}$ (Q = 108 C)	-

^aAverage current was found by dividing the charge transfer by the corresponding duration. Q, total charge transfer; IS, initial stage; RS, return stroke; CC; continuing current.

the order of 100 A [Rakov and Uman, 2003, p. 7, 272–274, 329–331]. Return strokes (impulsive current components) in rocket-triggered lightning are similar to subsequent return strokes in natural lightning [Fisher et al., 1993]. Both steady currents and return strokes produce NO_x, but the production by the latter is small compared to that by the former, as illustrated below.

[12] We can roughly estimate the contribution from return strokes to NO_x production using laboratory data of Wang et al. [1998], who employed 4-cm long sparks with lightning-like peak currents. They approximated the experimental dependence of the number of NO molecules per meter spark length on peak current (ranging from 7 to 29 kA) by a quadratic equation (see equation (6) and Figure 4 of Wang et al.). Note that in Wang et al.'s experiment 90–95% of the produced NO_x consisted of NO. Using this equation we estimate that the NO_x production by the three return strokes with peak currents 6, 34 and 12 kA in flashes 0510 and 0512 is about $5 \cdot 10^{21}$ NO_x molecules per meter, which is only about 20% of the total NO_x production, $2.4 \cdot 10^{22}$ NO_x molecules per meter, by flashes 0510 and 0512. It is clear that the main contribution came from steady currents. Since the steady current in triggered lightning is similar to the steady current in cloud discharges, the results presented here suggest that cloud discharges are as efficient as (or more efficient than) ground discharges in generating NO_x in the atmosphere.

[13] In-situ measurements of NO carried out in active thunderstorms, within the research project STERAO (Stratosphere-Troposphere Experiment: Radiation, Aerosols, and Ozone), suggest that lightning flashes produce about $2.0 \cdot 10^{20}$ to $1.0 \cdot 10^{22}$ NO molecules per meter [Stith et al., 1999]. In another study based on aircraft measurements of NO over central Europe, EULINOX (European Lightning NO_x Experiment), a mean value of $2.7 \cdot 10^{21}$ NO molecules per meter was estimated [Huntrieser et al., 2002]. In this latter study, the values for individual flashes span over 2 orders of magnitude. Skamarock et al. [2003] used data from the STERAO project in conjunction with interferometric channel lengths from Defer et al. [2003] and NO_x tracer transport models to obtain a NO_x yield of 10^{21} NO_x molecules per meter. Note that lightning originally produces NO, some of which is converted to NO₂, so that measurements of NO_x (mainly NO and NO₂) yield the originally produced amount of NO.

[14] DeCaria et al. [2005] used a three-dimensional cloud-scale chemical model to simulate NO production by lightning in a storm observed during STERAO-A field experiment. They found that a best match between the observed profile of NO in the storm anvil and the computed NO production is obtained when one assumes that the NO yield of an intra-cloud flash is 75 to 100% of the NO yield of a cloud-to-ground flash. Using field observations, Dye et al. [2000] also argued that cloud flashes are major contributors to NO_x production in thunderstorms. The results of the present study provide the first direct evidence that return strokes in cloud-to-ground flashes are not primary NO_x producers.

[15] It appears from the present study that NO_x production by lightning during the steady current phase is related to charge transfer, with the observed values being between 2 and $3 \cdot 10^{20}$ molecules per meter per coulomb.

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