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NO_x from lightning

1. Global distribution based on lightning physics

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Abstract. This paper begins a study on the role of lightning in maintaining the global distribution of nitrogen oxides (NO_x) in the troposphere. It presents the first global and seasonal distributions of lightning-produced NO_x (LNO_x) based on the observed distribution of electrical storms and the physical properties of lightning strokes. We derive a global rate for cloud-to-ground (CG) flashes of 20–30 flashes/s with a mean energy per flash of 6.7×10^9 J. Intracloud (IC) flashes are more frequent, 50–70 flashes/s but have 10% of the energy of CG strokes and, consequently, produce significantly less NO_x. It appears to us that the majority of previous studies have mistakenly assumed that all lightning flashes produce the same amount of NO_x, thus overestimating the NO_x production by a factor of 3. On the other hand, we feel these same studies have underestimated the energy released in CG flashes, resulting in two negating assumptions. For CG energies we adopt a production rate of 10×10^{16} molecules NO/J based on the current literature. Using a method to simulate global lightning frequencies from satellite-observed cloud data, we have calculated the LNO_x on various spatial (regional, zonal, meridional, and global) and temporal scales (daily, monthly, seasonal, and interannual). Regionally, the production of LNO_x is concentrated over tropical continental regions, predominantly in the summer hemisphere. The annual mean production rate is calculated to be 12.2 Tg N/yr, and we believe it extremely unlikely that this number is less than 5 or more than 20 Tg N/yr. Although most of LNO_x is produced in the lowest 5 km by CG lightning, convective mixing in the thunderstorms is likely to deposit large amounts of NO_x in the upper troposphere where it is important in ozone production. On an annual basis, 64% of the LNO_x is produced in the northern hemisphere, implying that the northern hemisphere should have natural ozone levels as much as 2 times greater than the southern hemisphere, even before anthropogenic influences. The amount of O₃ produced from this NO_x is expected to exceed the stratospheric source by a factor of 1.5, and thus the hemispheric asymmetry in LNO_x would lead to a significant excess of northern hemisphere O₃ even in the preindustrial troposphere. (The monthly climatologies for LNO_x on a $1^\circ \times 1^\circ$ latitude-longitude grid can be obtained by e-mail to cprice@flash.tau.ac.il.)

1. Introduction

Nitrogen oxides or NO_x (NO and NO₂) play a key role in the photochemical reactions which determine tropospheric and stratospheric ozone concentrations [Kroening and Ney, 1962; Crutzen, 1970; Johnston, 1971]. In the troposphere, regions with high NO_x concentrations produce O₃ both locally [Haagen-Smit, 1958] and globally [Chameides and Walker, 1973; Crutzen, 1979], while in those with low NO_x concentrations, O₃ is destroyed [Logan et al., 1981; Crutzen, 1983; Liu et al., 1983]. Because ozone is a greenhouse gas [Ramanathan and Dickinson, 1979; Lacis et al., 1990], knowledge of global NO_x concentrations is important for global climate studies. Radiative

forcing by O₃ is particularly sensitive to changes in the vertical distribution of O₃ in the upper troposphere, and hence it is important to understand the regional and vertical distributions of NO_x sources in the atmosphere. NO_x is also a key player in the determination of OH concentrations (both directly and through O₃ production) and thereby impacts the ability of the atmosphere to oxidize and remove many species [Levy, 1971; Logan et al., 1981]. Since NO_x is a short-lived chemical family and highly variable in the troposphere, it is extremely important to understand the regional source strengths of NO_x to properly understand tropospheric chemistry.

Lightning-produced NO_x (LNO_x) is primarily made up of NO and NO₂, with NO being 75–95% of the total [Franzblau, 1991]. When diluted with ambient air, the NO formed in the lightning channel reacts quickly with O₃ to form NO₂, and in daylight a photochemical balance between NO and NO₂ is established. Estimates of the global lightning-produced NO_x range from 1 Tg N/yr [Levine et al., 1981] to 100 Tg N/yr [Franzblau and Popp, 1989] (Tg = 10^{12} g). The other major sources of tropospheric NO_x are fossil fuel burning (~24 Tg

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N/yr), biomass burning (~8 Tg N/yr), soil emissions (~12 Tg N/yr), NH₃ oxidation (~3 Tg N/yr), aircraft (~0.4 Tg N/yr), and transport from the stratosphere (<0.4 Tg N/yr) [Prather and Logan, 1994; Houghton et al., 1995]. Of all the sources, lightning has the largest uncertainty. Better, more constrained estimates for LNO_x are needed, for example, to determine the relative role of the current aircraft fleet in enhancing NO_x and hence O₃ in the upper troposphere [Brasseur et al., 1996; Flato et al., 1995]. In part 1 of this study we present the first detailed global distributions of LNO_x as a function of latitude, longitude, and season. Part 2 derives an independent limit on LNO_x using the global atmospheric electric circuit. In part 3 the new lightning sources will be evaluated in different chemical tracer models, comparing the concentrations of LNO_x with that from aircraft and other sources.

Section 2 of this paper describes the methodology followed here. Section 3 discusses the various physical parameters needed to establish the available energy in lightning flashes. Section 4 addresses the amount of NO_x produced per unit energy in a lightning discharge. Section 5 presents the global distributions of lightning and NO_x produced using our methodology. Section 6 provides our discussion of the uncertainties involved in our calculations, and Section 7 presents our conclusions and the obvious, but not commonly discussed, implications for tropospheric ozone production.

2. Methodology

In order to calculate the global budget of LNO_x, one needs to answer three questions.

1. How much NO_x is produced per Joule of energy, and is this amount constant for all lightning energies? Estimates of the amount of NO_x produced per unit energy range anywhere from 2–2500 × 10¹⁶ molecules of NO per Joule [Borucki and Chameides, 1984]. These estimates are obtained from theoretical calculations [Tuck, 1976; Chameides et al., 1977; Griffing, 1977; Chameides, 1979; Hill et al., 1980; Goldenbaum and Dickerson, 1993], laboratory experiments [Chameides et al., 1977; Levine et al., 1981; Peyrous and Lapeyre, 1982], and atmospheric field observations [Nixon, 1976, 1978; Drapcho et al., 1983]. The reasons for the large uncertainty in this quantity are many.

Theoretical calculations have to make various assumptions regarding parameters of the lightning strokes, such as, temperature, density, peak current, conductivity, radius, and length of the channel. All of these parameters vary as a function of time during the lightning stroke and can vary from one lightning stroke to another, producing a distribution of energies in lightning strikes. Chameides [1979] indicates that the amount of NO_x produced by a lightning discharge is not a linear function of the discharge energy. This implies that using a single value for the amount of NO_x produced per Joule is probably incorrect. Furthermore, the typical calculations of NO_x production in lightning discharges [Zel'dovich and Raizer, 1966] do not consider the effects of water vapor on the production of NO_x. Peyrous and Lapeyre [1982] have shown that the NO_x production is strongly dependent on relative humidity.

Laboratory experiments have a basic scaling problem. Sparks created in the laboratory have very small peak currents relative to natural lightning discharges. This implies that the energies in these small discharges are much lower than those found in lightning channels. Each experiment that is slightly different produces different results. Even if the NO_x produced

per unit energy in the lab is correct, can one extrapolate these values to a lightning stroke that has peak temperatures of 30,000 K [Orville, 1968] and pressures of possibly 50 atm [Hill, 1971]? Furthermore, NO_x produced by the corona sheath surrounding the lightning channel has been speculated to produce 50% of the total NO_x. This NO_x would only be produced when large electric fields are present around the lightning channel. This does not occur in small scale laboratory experiments.

Field measurements have their problems too. In addition to difficulties with accurately integrating the NO_x produced in a flash (and dispersed in the storm), all field measurements calculate the concentration of NO_x per flash (not per joule). Since flashes can have a great range of peak currents, number of return strokes, channel lengths, and hence energies, these observations tell us little about the NO_x produced per unit energy. To do this, assumptions have to be made regarding the above parameters. Furthermore, since it appears that intra-cloud (IC) flashes have significantly lower energies than cloud-to-ground (CG) discharges [Holmes et al., 1971; Kowalczyk and Bauer, 1981], it is incorrect to divide the total NO_x observed over a certain period by the total number of flashes.

2. How much energy is available per flash? The values quoted in the literature range from 10⁸ to 10¹⁰ J per flash [Chalmers, 1967; Uman, 1987]. The reason for this range is that the energies in lightning flashes are related to the peak currents in lightning discharges, which vary by 2 orders of magnitude [Uman, 1987]. Therefore a single observed or calculated value may not be representative of the mean value. The majority of previous studies have used a single value for their global calculations, and this value differs from paper to paper. The mean value often used is 4 × 10⁸ J/flash [Chameides, 1986; Lawrence et al., 1995] and is derived from an analysis of optical energies radiated from lightning channels. However, the optical energy is estimated to be only 0.5% of the total energy [Connor, 1967; Krider et al., 1968; Borucki and Chameides, 1984]. The conversion from optical to total energy therefore introduces large uncertainties. Furthermore, the optical energies used to derive the total energy were obtained in large part from the optical lightning observations from the DMSP satellite [Turman, 1978]. Not only did these observations primarily observe IC flashes, which are significantly less energetic than CG flashes [Holmes et al., 1971; Kowalczyk and Bauer, 1981], but the detection efficiency of the lightning sensor aboard the DMSP satellite is thought to be only 2% [Turman and Edgar, 1982], thereby resulting in observations of only the brightest flashes.

There are other studies that calculate the energy in units of joules per meter, derived from electric field measurements. Even if these electric field measurements are correct, to get the total energy in a lightning flash, one has to make assumptions regarding the length of the lightning channel. Although the CG length can range from 5 to 7 km [Krehbiel, 1986], the IC length is 1–6 km [Ogawa and Brook, 1964; Krehbiel, 1986]. Furthermore, no one has considered tortuosity in their calculations. A lightning channel is not a straight line. It is possible that the length of a channel could be increased by at least a factor of 2 if it was stretched out like a piece of string (V. Idone, personal communication, 1993). This would result in the previous estimates of total energy being doubled.

3. How does one extrapolate to get a global estimate of NO_x production by lightning? Even if values obtained to answer questions 1 and 2 are correct, one then needs to extrapolate these values to get a global number for the NO_x produc-

tion by lightning. The single biggest simplification made by the vast majority of researchers in this field is to assume that globally, there are 100 equally efficient NO_x-producing flashes per second [Noxon, 1976; Tuck, 1976; Chameides et al., 1977; Griffing, 1977; Dawson, 1980; Hill et al., 1980; Levine et al., 1981; Drapcho et al., 1983; Peyroux and Lapeyre, 1982; Borucki and Chameides, 1984; Bhetanabhotla et al., 1985; Franzblau and Popp, 1989; Liaw et al., 1990; Sisterton and Liaw, 1990; Lawrence et al., 1995]. This magical number of 100 was originally an order of magnitude estimation [Brooks, 1925]. Brooks arrived at this estimation by combining the results of his climatological survey of thunderstorm frequencies with the lightning frequencies obtained by Marriott [1908] during a single 28-min observation period of a single thunderstorm in West Norwood, England, on June 4, 1908. It is remarkable how influential this single observation has been in the scientific literature. It was later confirmed by satellite observations that Brooks' estimation was of the correct order of magnitude [Orville and Spencer, 1979; Turman and Edgar, 1982; Kotaki and Katoh, 1983]. However, the global values ranged from 40 to 140 flashes per second, and these values were for total lightning frequencies (IC + CG). However, IC flashes appear to have energies much lower than CG flashes [Holmes et al., 1971; Kowalczyk and Bauer, 1981; Sisterton and Liaw, 1990] and therefore do not play a major role in the production of NO_x. Since CG flashes make up approximately 30% of the total flashes [Mackerras and Darveniza, 1994], all global estimates using 100 flashes/s should be scaled down by a factor of 3! Some researchers acknowledge that IC flashes are more frequent than CG flashes, however, they multiply by 3 instead of dividing by 3 [Tuck, 1976; Chameides et al., 1977; Peyroux and Lapeyre, 1982] to obtain the number of NO_x-producing flashes.

In addition to the above simplification, lightning energies may vary greatly around the globe, possibly being greater toward the tropics. Orville [1990] has shown that even within the United States there appears to be a latitudinal gradient in the lightning peak currents. He showed that Florida thunderstorms had peak currents nearly double those found in New England. However, Petersen and Rutledge [1992] have shown that the mean peak currents in CG flashes in the tropics were very similar to the values found in Florida. Finally, the ratio of IC/CG has been shown to vary as a function of latitude [Pierce, 1970; Price and Rind, 1993; Mackerras and Darveniza, 1994], which also needs to be considered when making global estimations of LNO_x.

The above three major areas of uncertainty result in the large range of global estimates often quoted for the production of NO_x by lightning. Extensive work has already been carried out regarding uncertainty 1, and we feel we cannot reduce this uncertainty. Thus we have focused our efforts on the remaining uncertainties 2 and 3.

3. Physics of Lightning

To understand the energies involved in lightning discharges, one has to consider the various lightning parameters that are present in a typical thunderstorm.

Breakdown Potential

It is still debatable as to what mechanism results in the electrification of clouds [e.g., Saunders, 1994]. Whatever the mechanism, it is observed that a "typical" thunderstorm is a positive dipole, with a net negative charge in the lower parts of

the cloud and a net positive charge in the upper parts of the cloud [Wilson, 1920; Simpson and Scrase, 1937; Williams, 1985]. The charge centers in thunderstorms occur at temperatures below 0°C and above -40°C. This points to the importance of ice and supercooled droplets in the electrification process. In fact, the volume of the mixed-phase region is thought to be related to the intensity of lightning activity in thunderstorms [Price and Rind, 1993]. The negatively charged region near the base of the cloud induces a positively charged region on the ground below, resulting in strong electric fields developing between the cloud and the Earth [Standler and Winn, 1979]. When the electric field reaches a threshold, or breakdown potential, a lightning discharge occurs. The discharge can occur between the cloud and the ground (cloud-to-ground flash) or within the cloud (intracloud flash). Occasionally, discharges also occur between clouds (intercloud flash) or upward from cloud tops into the stratosphere (cloud-to-air flash).

The maximum electric fields in thunderstorms were studied extensively by Winn et al. [1974]. Although they found that the field could be as high as 4.5×10^5 V/m, the mean value was approximately 5×10^4 V/m. Considering that the negative charge center in the cloud is between 5 and 7 km above the ground [Krehbiel, 1986], and the length of an intracloud (IC) discharge is between 1 and 6 km [Ogawa and Brook, 1964; Krehbiel, 1986], a breakdown potential of $2.5\text{--}3.5 \times 10^8$ V for CG discharges and $0.5\text{--}3 \times 10^8$ V for IC discharges is expected. These values are in good agreement with previously published values [Malan, 1963; Tzur and Roble, 1985; Borovsky, 1996].

Cloud-to-Ground Lightning

There are two basic types of CG flashes: those that bring negative charge to the Earth's surface and those that bring positive charge to the Earth's surface. Because of the proximity of the negative charged region in the cloud to the Earth, most CG flashes are negative flashes [Uman and Krider, 1989; Petersen and Rutledge, 1992; Orville, 1994]. It should be noted that this value can vary considerably from storm to storm [Beasley, 1985] and varies as a function of season (K. Cummins, personal communication, 1995).

A typical discharge begins within the cloud with a process called preliminary breakdown. This lasts for tens of milliseconds and initiates an intermittent, highly branched discharge, called a stepped leader. As the downward propagating leader comes close to the ground, an upward propagating discharge, or streamer, is initiated from the ground. When the two discharges meet, a large current pulse, known as the return stroke, starts at the ground and propagates back up the previously ionized leader channel to the cloud. If additional cloud charge is available at the end of the first return stroke, a dart leader propagates, without stepping, down the previous return stroke channel and initiates a subsequent stroke. The strokes are often sufficiently separated in time to produce a flickering effect to the observer. A lightning flash is made up of a number of strokes. NO_x is produced primarily during the high-energy return stroke phase of the flash and not during the leader phase (see section 4).

Peak current statistics of CG flashes have been analyzed using data from the U.S. National Lightning Detection Network (NLDN) [Orville, 1991a] for the period June–August 1988 (R. Orville, personal communication, 1994). The network observes only CG flashes, supplying information on polarity, peak currents, and multiplicity (number of return strokes). It should be noted that only the peak current from the first return

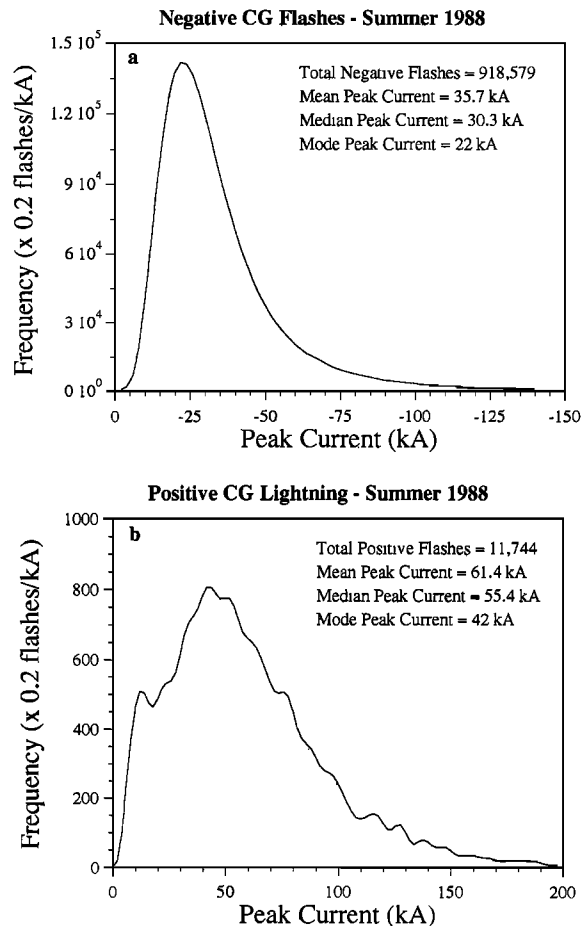


Figure 1. Frequency distribution of (a) negative and (b) positive cloud-to-ground lightning flashes observed by the U.S. National Lightning Detection Network in the United States during the summer of 1988.

stroke is recorded. Furthermore, the peak current is obtained from the peak signal strength using the calibration published by Orville [1991b]. The normalized (to 100 km) peak current is obtained by multiplying the peak signal strength by 0.2. Because of the tremendous amount of data, only 14 days during the above period were analyzed, each day separated by 1 week (June 1, 8, 15, 22, 29, July 6, 13, 20, 27, and August 3, 10, 17, 24, 31). For the 14 days (24 hours per day) there were 930,323 cloud-to-ground lightning flashes detected by the network. Of these ground flashes, 918,579 (98.7%) were negative flashes.

The frequency distribution of the peak currents in negative and positive flashes is shown in Figure 1a and 1b. The mean negative flash peak current is 35.7 kA, while the mean positive flash peak current is 61.4 kA. The median negative peak current is 30.3 kA, in excellent agreement with observations from other locations around the globe [Malan, 1963; Berger *et al.*, 1975; Uman and Krider, 1989; Petersen and Rutledge, 1992].

The mean peak current in a thunderstorm is strongly dependent on the sample size used for averaging. To indicate the dependence of peak current on lightning frequency in a particular thunderstorm, the negative flash data for the United States were binned into grid boxes of $1^\circ \times 1^\circ$ (longitude \times latitude) and averaged over time intervals of 15 min. The mean values of the observed parameters were only used in this analysis if the frequency of lightning in a grid box was greater than

1 CG flash per minute (i.e., at least 15 flashes used for each average).

The mean peak negative currents as a function of negative flash frequency are shown in Figure 2a. Although there is a large amount of variability in mean peak currents, as the sample size increases (high flash frequencies), the mean peak current converges on a value of approximately 35 kA. The multiplicity can be represented in a similar fashion (Figure 2b). Mean values for the multiplicity can range from 1 to 6 return strokes per flash. However, as the frequency of the lightning increases in the storm, the mean multiplicity converges on a value of three return strokes per CG flash. In agreement with previous findings we find that the vast majority (87%) of positive flashes in this data set have only one return stroke.

Since the charge deposited by a return stroke is difficult to measure, one can calculate the charge transferred to the Earth by integrating over the current pulse ($Q = \int I dt$). Various models for the return stroke current pulse have been proposed [Bruce and Golde, 1941; Barlow *et al.*, 1954; Hepburn, 1957; Gosh and Khastgir, 1972]. The general analytical expression that best matches the observed values of lightning currents is represented by

$$I(t) = I_0[Ae^{(-\alpha t)} - Be^{(-\beta t)} + Ce^{(-\gamma t)}] \quad (1)$$

where I_0 = peak current. Constants α and β determine the rise and decay time to and from the peak current value. These values are well observed. For the first return stroke the mean risetime to the peak current is approximately $5 \mu\text{s}$, with a decay time to half the peak current of $40\text{--}50 \mu\text{s}$ [Berger *et al.*, 1975;

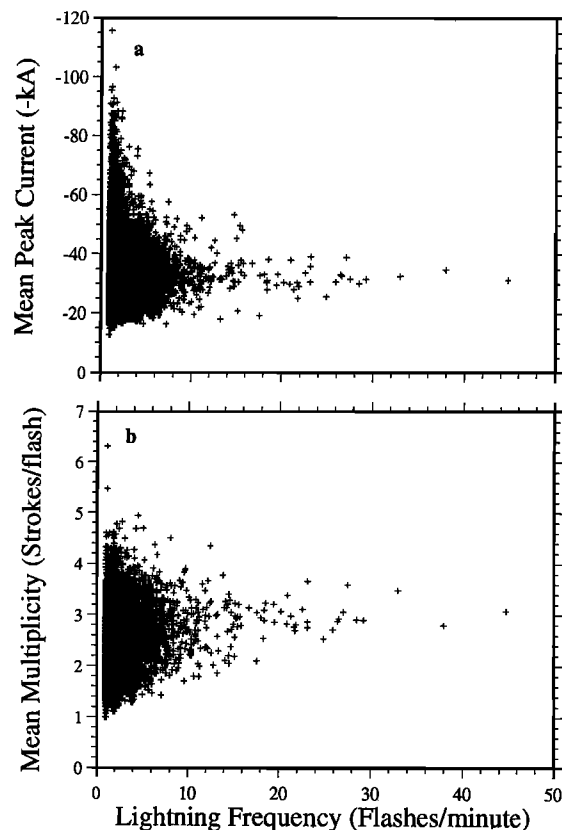


Figure 2. (a) Mean peak current and (b) mean multiplicity of cloud-to-ground lightning as a function of lightning frequency in thunderstorms.

Uman, 1987]. These timescales result in values of $\alpha = 3.3 \times 10^4/s$ and $\beta = 4.5 \times 10^5/s$. Subsequent return strokes have risetimes of $1 \mu s$ and similar decay times resulting in $\alpha = 2.5 \times 10^4/s$ and $\beta = 3.8 \times 10^6/s$. The third term in (1) represents the low-intensity current tail. Barlow *et al.* [1954] and Hepburn [1957] obtained values for γ by fitting the equation to observations of return strokes. They found values of $\gamma = 10.1 \times 10^2/s$ and $7 \times 10^2/s$, respectively. We use a value of $8.8 \times 10^2/s$, an average of these two values. For the first return stroke, $A = B = 1$ [Bruce and Golde, 1941] and $C = 0.25$, an average of the values found by Barlow *et al.* [1954] ($C = 0.3$) and Hepburn [1957] ($C = 0.2$). Subsequent strokes have a peak current approximately half that of the first stroke, therefore for these strokes, $A = B = 0.43$ and $C = 0.11$ (or 0.25×0.43). The return strokes can therefore be represented by

First return stroke

$$I(t) = I_0[e^{-3.3 \times 10^4 t} - e^{-4.5 \times 10^5 t} + 0.25e^{-8.8 \times 10^2 t}] \quad (2)$$

Subsequent return stroke

$$I(t) = 0.43I_0[e^{-2.5 \times 10^4 t} - e^{-3.8 \times 10^6 t} + 0.25e^{-8.8 \times 10^2 t}] \quad (3)$$

Using our mean observed negative peak current of $I_0 = 35.7$ kA (Figure 1a), the current profiles are shown in Figure 3 for the first and subsequent return strokes during the first $100 \mu s$. It should be noted that up to 50% of CG flashes have return strokes ending with a long period of low-intensity current known as continuing current [Uman and Krider, 1989]. This continuing current has a mean amplitude of 150 A lasting for approximately 150 ms [Ogawa, 1982].

To obtain the charge (Q) deposited by the first and subsequent return strokes, one simply integrates $I(t)$. This results in

First return stroke

$$Q_1 = 3.12 \times 10^{-4} I_0 \quad (4)$$

Subsequent return stroke

$$Q_s = 1.39 \times 10^{-4} I_0 \quad (5)$$

The curves in Figure 3 ($I_0 = 35.7$ kA) result in a charge transfer of 11.1 C for the first return stroke and 5 C for the subsequent strokes. These values compare favorably with observed values [Berger *et al.*, 1975; Uman, 1987]. Strokes with continuing current have an additional charge transfer of $Q = 150 \text{ A} \times 150 \times 10^{-3} \text{ s} = 22.5 \text{ C}$. The majority of charge deposited by a lightning flash therefore occurs during the continuing current phase of the flash. However, the continuing current period does not appear to be of importance for NO_x production (see section 4), and the NO_x production in this study will be approximated using return strokes without continuing current.

Intracloud Lightning

The vast majority of lightning occurs within clouds and not between the clouds and the ground [Pierce, 1970; Prentice and Mackerras, 1977; Price and Rind, 1993; Mackerras and Darveniza, 1994]. As mentioned previously, the electrification of thunderstorms occurs in the mixed-phase region of clouds (between 0°C and -40°C) resulting from interactions between supercooled droplets and ice particles. As the electrification in the mixed-phase region intensifies, the probability of breakdown in the cloud increases. Thunderstorms with larger volumes above the freezing level (0°C isotherm) generally tend to

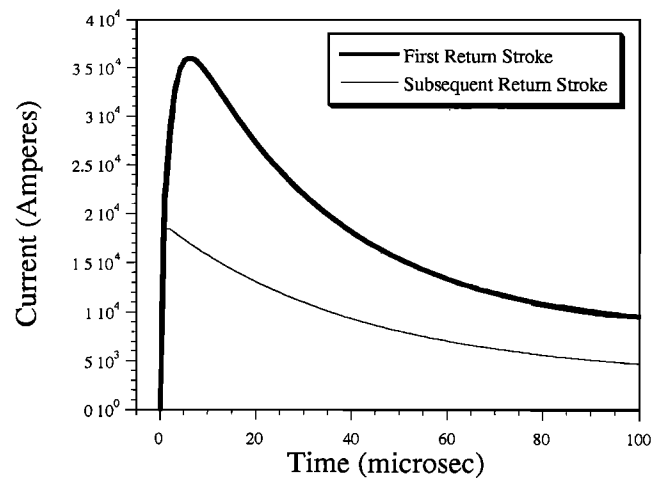


Figure 3. Temporal development of the current flowing through a lightning channel during the first and subsequent return stroke.

have more IC flashes relative to CG flashes [Price and Rind, 1993], although the proportion of CG flashes in an individual thunderstorm can be highly variable. Since tropical thunderstorms are often deeper than midlatitude storms, this may explain why the IC/CG ratio is observed to be larger in the tropics than in midlatitudes [Pierce, 1970; Prentice and Mackerras, 1977].

The characteristics of an IC flash are somewhat different from those of a CG flash. The IC discharge occurs between two or more diffuse centers of opposite charge. For IC flashes, the leader stage of the discharge is the principal process by which charge is transferred within the cloud to neutralize the charge centers. During the leader stage, currents of the order of 100 A flow through the IC channel. However, as the leader progresses, it occasionally encounters localized concentrations of opposite charge. This results in a rapid increase in the current from a few hundred amperes to a few thousand amperes. These pulses are known as K changes and have been estimated to transfer 3.5°C within a few milliseconds, resulting in average currents of 1–4 kA [Ogawa and Brook, 1964]. The observed electric field changes associated with K changes are very similar to the field changes caused by CG return strokes [Kitagawa and Brook, 1960]. As with the CG return strokes, we believe that the majority of NO_x is produced during the rapid expansion and cooling of the hot channel (see section 4), which for the intracloud discharge occurs during the K changes.

Lightning Energies

The energy (E) in a lightning flash (stroke) can be determined in two ways:

$$E_1 = L \int \frac{I(t)^2}{\sigma(t) \pi r(t)^2} dt \quad (\text{joules}) \quad (6)$$

$$E_2 = V \int I(t) dt = VQ \quad (\text{joules}) \quad (7)$$

where $I(t)$ represents the current pulse, $\sigma(t)$ is the electrical conductivity of the lightning channel, $r(t)$ is the radius of the channel, V is the breakdown potential, and L is the length of the lightning channel. The conductivity is strongly dependent

on the lightning channel temperature. Peak temperatures between 30,000–40,000 K are reached within a few microseconds [Orville, 1968]. The electrical conductivity varies by 11 orders of magnitude as the temperature rises from 2000 K to 30,000 K [Yos, 1963; Uman and Voshall, 1968]. Furthermore, the channel radius varies by 2 orders of magnitude, from 1 mm to 10 cm, during the lifetime of the flash. Both these parameters are highly uncertain, resulting in large uncertainties in E_1 . In addition, to use E_1 , an additional assumption has to be made regarding the channel length, L , before calculating the total energy.

Using the second formulation for energy (E_2), one only has to assume a value for the breakdown potential (V) which does not vary over the lifetime of a lightning flash and is better known than $\sigma(t)$, $r(t)$, and L . The breakdown potential can vary from storm to storm but is fairly constant during a particular storm. As clouds become increasingly electrified during a storm's development, breakdown simply occurs more often, resulting in larger lightning frequencies. However, the value of the breakdown potential remains fairly constant throughout the storm. For the above reasons we have decided to use E_2 for the calculation of the energies in lightning flashes.

From Figure 2b we adopt a mean value of 3 for the multiplicity of negative flashes. Therefore using (4) and (5), we get

$$Q_- = Q_1 + 2Q_s = 5.9 \times 10^{-4} I_0 \quad (8)$$

For the breakdown potential ($2.5\text{--}3.5 \times 10^8$ V) we adopt a mean value of 3×10^8 V, giving an energy of

$$E_- = Q_- V = 1.77 \times 10^5 I_0 \quad (9)$$

For mean negative peak currents ranging from 10–60 kA (90% of data used in Figure 1a), the above implies a flash energy of $1.8 \times 10^9\text{--}1.1 \times 10^{10}$ J, in good agreement with previously published values [Uman, 1987].

Positive CG lightning flashes have mean peak currents of 61.4 kA (Figure 1b). However, approximately 90% of all positive flashes have only one stroke, unlike the mean value of three strokes in the negative flashes. Assuming positive flashes have the same temporal evolution as negative flashes (Figure 3), the charge deposited by a mean positive flash, neglecting the continuing current portion of the stroke, is slightly less than that for a mean negative flash [$Q_+ = 0.87Q_1 + 0.13(Q_1 + Q_s) = 0.96Q_-$]. The breakdown potential for positive flashes is not well known, although considering that positive flashes normally originate from the anvils of thunderstorms (approximately 10 km), the breakdown potential can be assumed to be approximately 5×10^8 V (5×10^4 V/m \times 10000 m). This is a factor of 1.67 larger than for a negative flash. Therefore the energy in a positive flash (E_+) is assumed to be a factor of 1.6 (1.67×0.96) larger than that for a negative flash. Although only a small fraction of the total lightning is represented by positive flashes (1.3% in this study), it has been shown that in the United States between 1989 and 1991, 3.7% of all CG flashes were positive [Orville, 1994], while in the tropics the fraction may be as large as 10% [Petersen and Rutledge, 1992]. We therefore assume that on a global scale, positive lightning accounts for approximately 5% of the total CG lightning. Therefore the weighted energy per CG flash is

$$\begin{aligned} E_{CG} &= 0.95E_- + 0.05E_+ = 0.95E_- + 0.05(1.6E_-) \\ &= 1.03E_- = 1.823 \times 10^5 I_0 \end{aligned} \quad (10)$$

Using the mean peak current for negative and positive CG flashes, the weighted mean energy for a CG flash is

$$\begin{aligned} E_{CG} &= 1.823 \times 10^5 (0.95(35.7 \times 10^3) + 0.05(61.4 \times 10^3)) \\ &= 6.7 \times 10^9 \text{ J} = 6.7 \text{ GJ} \end{aligned} \quad (11)$$

with a range of 1.8–11 GJ deduced from the range of peak negative currents of 10–60 kA and peak positive currents ranging from 10 to 100 kA (Figure 1).

The breakdown potential for IC flashes is uncertain but cannot be larger than that for CG flashes. In fact, the breakdown potential is probably less due to the smaller distances between the charge centers [Ogawa and Brook, 1964] and because of the presence of water droplets and ice within the cloud that reduce the electric fields [Mackay, 1931]. The cloud particles produce localized electric fields much larger than the mean field and effectively reduce the breakdown potential. Nevertheless, as presented earlier, the breakdown potential for intracloud flashes ($0.5\text{--}3 \times 10^8$ V) is taken as 1.75×10^8 V, while the charge transferred in a K change is taken as 3.5°C [Ogawa and Brook, 1964]. Therefore from equation (7), the energy for K changes is 6.1×10^8 J, at least an order of magnitude less than that found in CG return strokes. This is in agreement with estimations by Kowalczyk and Bauer [1981] and Sisterson and Liaw [1990] who assumed IC flashes to have one tenth the energy of CG flashes. Holmes et al. [1971], studying the thunder signatures of lightning, found that IC flashes had one third the energy of CG flashes. However, the acoustic energies ranged from 1 to 17×10^6 J, which is only 0.1% of the observed total energies. Therefore it is difficult to arrive at a ratio between the total energies of IC and CG flashes from these measurements. As described above, we believe the majority of NO_x is produced during the K-change process of the intracloud flash. Therefore we simply adopt a value of $E_{IC} = 0.1 E_{CG}$ for the available energy in the rapidly expanding hot channel, however, the ratio could well be smaller.

4. Production of NO_x in the Lightning Channel

The high temperatures and high pressures initiated by a lightning stroke produce a shock front that cools as it expands outward. Nitric oxide (NO), produced by the Zel'dovich mechanism, is expected to have volume mixing ratios of 1–4% when the air cools to 2000 K–3000 K [Zel'dovich and Raizer, 1966]. This mechanism considers the breakup of N₂ and O₂ molecules in the hot lightning channel and the subsequent formation of NO. The final yield of NO is determined by the rate of cooling, the density of air behind the shock front, the radius of the channel, and the ambient pressure [Chameides, 1979; Goldenbaum and Dickerson, 1993].

NO is produced as the shock front propagates outward through a volume of air. The peak current in the discharge determines the energy deposited in the channel and hence the volume of air processed by the shock front, or the maximum radius of the channel. As the channel cools from outside inward, the amount of NO produced at each point is governed by the rate of cooling. The rate of cooling together with the volume of air processed determines the final NO production per flash. Stepped and dart leaders may also contribute to the NO_x production; however, the radius of leaders is at maximum a few millimeters, at least an order of magnitude less than the final radius of the return stroke [Borovsky, 1996], implying 2 orders of magnitude less in the volume of air processed by the leaders. The leaders are therefore assumed to produce much less NO_x as a result of the Zel'dovich mechanism. In addition,

during the continuing current stage of the discharge, temperatures can rise above the threshold for NO_x formation; however, the continuing current phase always follows the initial return stroke that heats the channel to 30,000 K. Since all N₂ and O₂ in the channel are already dissociated before the onset of the continuing current, any additional input of energy from the continuing current does not result in any further dissociation of N₂ and O₂. Furthermore, while the channel is expanding outward, the temperature behind the shock front does not drop substantially [Hill, 1970; Tuck, 1976; Hill et al., 1980]. Therefore the continuing current period also does not appear to be of importance for NO_x production. Corona discharges around the lightning channel and within the thunderstorm cloud may add to the production of NO_x; however, it is believed that the production of NO_x by corona discharge is a few orders of magnitude less than the production of NO_x by the hot channel [Bhetanabhotla et al., 1985; Sisterton and Liaw, 1990].

Direct observations of the amount of NO_x produced by lightning is difficult. Enhanced concentrations of nitrogen oxides have been observed within thunderstorms. Noxon [1976] observed NO₂ produced by a thunderstorm and derived a production rate of 2×10^{26} molecules of NO₂ per flash. The yield of NO_x molecules in a lightning stroke could, in principle, be determined from observations. Unfortunately, the number and energies of lightning strokes in a storm are not easy to measure, and it is even more difficult to integrate over the air entering and leaving a thunderstorm in order to quantify the addition of NO molecules or their subsequent chemical products such as NO₂. Observations that exist may be useful in bounding the problem, but careful propagation of all of the uncertainties places only loose constraints on P , the yield of NO molecules per joule. In addition to uncertainties in the primary production rate, if we are measuring only NO₂ we must also know what fraction of the NO produced has reacted with O₃.

Laboratory measurements and theoretical models remain the best approach to estimating NO_x production in lightning and related discharges. The key variable appears to be the energy density and the radius of the core channel. Observations of low-energy sparks in chambers [Chameides et al., 1977; Levine et al., 1981; Peyroux and Lapeyre, 1982] show that fewer NO molecules per joule of energy are produced in lower-energy discharges, and this is consistent with simple theoretical models [Chameides, 1979; Goldenbaum and Dickerson, 1993]. At the upper limit of energy density is the fireball in a nuclear weapon's test, which is also predicted to have low yields of NO per joule, while the energy densities occurring in lightning discharges span the peak yield of NO [Chameides, 1979].

Although Borucki and Chameides [1984] argue that P can be derived from theory with small uncertainty, $9 \pm 2 \times 10^{16}$ molecules NO/J, the recent work of Goldenbaum and Dickerson [1993] points out that both energy density and ambient pressure control the expansion of the shock front. Hence the yield P is very sensitive to the energy as well as pressure, with a narrow region of maximum P of 25×10^{16} molecules NO/J and a broad domain of $10\text{--}15 \times 10^{16}$ molecules NO/J at higher-energy densities ($5\text{--}10$ MJ/m³) more typical of CG strokes. Note that the average stroke considered by Goldenbaum and Dickerson [1993] (0.6 GJ) is much smaller than the estimate for the first return stroke here (3 GJ). At lower atmospheric pressures the energy typical of lightning strokes is predicted to produce very little NO [Goldenbaum and Dickerson, 1993]. Both of these analyses and the laboratory experiments point to

a maximum production in NO of the order of 10×10^{16} molecules NO/J in CG lightning.

Recently, upper atmospheric discharges have been discovered above thunderstorms at altitudes of 50–100 km [Sentman and Wescott, 1993; Lyons, 1994]. According to the Zel'dovich mechanism for NO production these discharges (known as "sprites") would produce little NO due to their low-energy densities and due to the low ambient pressure [Goldenbaum and Dickerson, 1993]. Furthermore, on a global scale these sprites are much less frequent than regular lightning and therefore are not expected to contribute to the global production of LNO_x.

We feel that the uncertainty in the mean production of NO, which includes also the uncertainties in averaging over the range of lightning-stroke energy densities and altitudes, is probably the largest uncertainty in NO production from lightning. We select a best value for P of 10×10^{16} molecules NO/J, with a bounding range of $5\text{--}15 \times 10^{16}$ molecules NO/J.

5. Global Lightning and NO_x Distributions

The lightning parameterizations used to approximate global lightning activity have been developed and described by Price and Rind [1992a, 1992b, 1993]. These are based on observations that show that thunderstorm electrification is closely linked to the updraft intensity in thunderstorms, which in turn is linked to the vertical development of convective clouds. For total lightning frequencies, two formulations are used: one for continental thunderstorms and one for oceanic thunderstorms, both using convective cloud top height as the predictive variable. The continental parameterization is based on observations showing that lightning frequencies in continental thunderstorms are related to approximately the fifth power of the cloud height [Shakford, 1960; Jacobson and Krider, 1976; Livingston and Krider, 1978; Williams, 1981, 1985]. The parameterization for marine thunderstorms is based on observations that show marine thunderstorms having very weak updraft intensities [Jorgensen and LeMone, 1989], resulting in extremely low lightning frequencies in these thunderstorms [Takahashi, 1990]. The relationships used are

$$F_c = 3.44 \times 10^{-5} H^{4.92} \quad (12)$$

$$F_m = 6.40 \times 10^{-4} H^{1.73} \quad (13)$$

where F_c and F_m are the lightning frequencies for continental and marine thunderstorms, respectively, (flashes per minute), and H is the cloud top height above ground (km).

To calculate the cloud-to-ground lightning frequencies, a method has been developed to calculate the proportion of cloud-to-ground flashes in a thunderstorm [Price and Rind, 1993]. The method relates the fraction of CG lightning in a storm to the thickness of the cold cloud portion of the cloud (0°C to cloud top). As the mixed-phase region within the cold sector expands as a cloud develops, the probability of dielectric breakdown increases due to increases in the electric field strength in this region. This results in a larger increase in intracloud lightning discharges in the thunderstorm relative to cloud-to-ground discharges. Therefore as the thickness of the cold cloud sector increases, the fraction of cloud-to-ground flashes decreases [Rutledge et al., 1992; Price and Rind, 1993]. The empirically derived formulation used to determine the proportion of cloud-to-ground flashes (PG) in an individual thunderstorm is

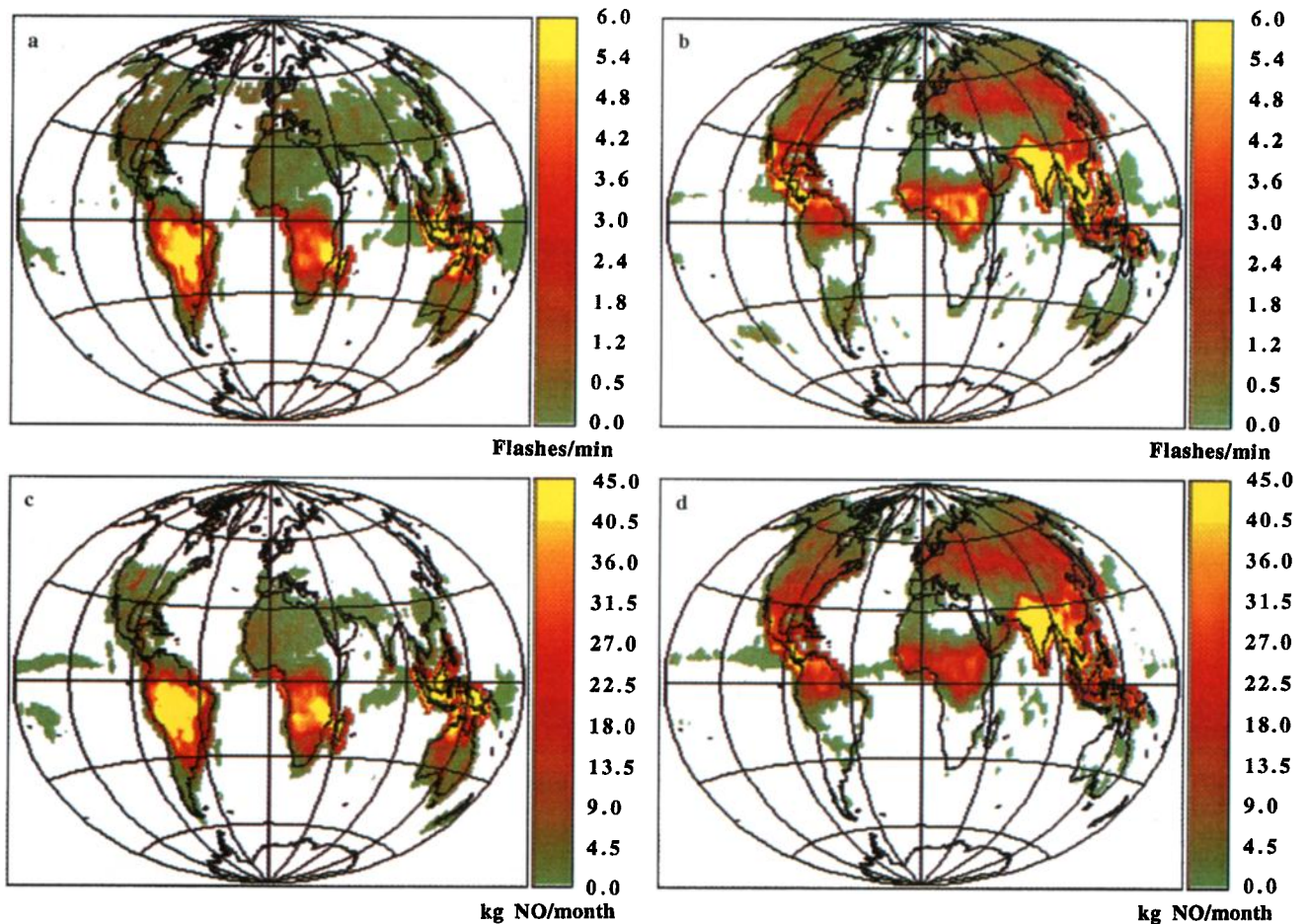


Plate 1. Global lightning distributions for (a) January 1988 and (b) July 1988 based on International Satellite Cloud Climatology Project cloud data. (c and d) Equivalent monthly mean LNO_x distributions for these two months.

$$PG = (aT^4 + bT^3 + cT^2 + dT + e)^{-1} \quad (14)$$

where $a = 0.021$, $b = -0.648$, $c = 7.49$, $d = -36.54$, $e = 64.09$, and $T =$ cold cloud thickness (km). This formulation was developed only for cold cloud thicknesses greater than 5.5 km, and less than 14 km [Price and Rind, 1993]. When $T < 5.5$ km, very little lightning occurs (see equations (8) and (9)). In this case, it is assumed that all lightning is intracloud and no cloud-to-ground lightning occurs. This is what is observed in the initial developmental stage of thunderstorms. For $T > 14$ km most of the lightning occurs as intracloud flashes, and the proportion of CG flashes is taken as being equal to that for $T = 14$ km, or 2%. To get the total number of CG flashes, the fraction of CG flashes in a storm (PG) is multiplied by the total lightning frequency in the thunderstorm, derived using (8) and (9).

The global lightning frequencies were calculated using cloud data from the International Satellite Cloud Climatology Project (ISCCP), which provides global convective cloud data at three hour intervals from July 1983 to June 1991 [Rossow and Schiffer, 1991]. The initial spatial resolution of the data is approximately 5 km, fine enough to resolve individual thunderstorms. Thunderstorm clouds are identified by their cloud top temperatures (infrared radiances) and their optical depths (visible radiances). Because of uncertainties in determining cloud types from satellites at high latitudes (high solar zenith

angle), the cloud data at solar zenith angles greater than 85° were not used in our calculations. This usually occurs at high latitudes in the winter hemisphere. Furthermore, since optical depth values can only be obtained during daylight hours, the following analysis represents daytime lightning and NO_x concentrations only. However, observations of global lightning distributions at local midnight [Orville and Henderson, 1986] show great similarity to observations of global lightning taken at local dawn and dusk [Turman and Edgar, 1982]. Furthermore, Olapido and Mornu [1985] have shown that for a tropical station in Nigeria (11°N), a 14-year study revealed that 43% of the lightning occurred between 0600 and 1800 LST, while observations from Darwin, Australia (11°S), show a similar division between daytime and nighttime lightning [Williams and Heckman, 1993]. Until observations of the local diurnal cycle of lightning are made on a global scale, we cannot know if using only daytime lightning frequencies will provide a systematic bias to our results.

Using the International Satellite Cloud Climatology Project (ISCCP) data set, the simulated total lightning frequencies for January and July 1988 are shown in Plates 1a and 1b. The two main features of the global lightning distributions are the concentration of lightning primarily over continental regions while mainly in the summer hemisphere. Lightning activity migrates north and south of the equator with the changing seasons.

Integrating over the globe, we get a total of 71 flashes/s during January 1988 and 101 flashes/s in July 1988. During January the cloud-to-ground frequency is 19 flashes/s, while during July, it is 30 flashes/s. Comparisons between simulated and observed global lightning distributions show good agreement [Price and Rind, 1992a, b; Price, 1993].

Given the previously calculated mean energy per flash (6.7×10^9 J), it is now possible to calculate the regional and global NO_x production (G) per month.

$$G = (CG \cdot E_{CG} + IC \cdot E_{IC})tPC = (CG + 0.1 IC)(E_{CG})tPC \quad (15)$$

where CG is the cloud-to-ground flash frequency (flashes/second), IC is the intracloud flash frequency, E_{CG} is the mean energy per CG flash (J) and 0.1 is the ratio of energies for IC and CG flashes, t is the period of interest (day, month, year) in seconds, P is the production rate (molecules NO_x/J), and C is a conversion factor equal to $(14 \text{ g/mole})/(6.02 \times 10^{23} \text{ molecules/mole})$ or $2.33 \times 10^{-23} \text{ g/molecule}$. The regional distribution of LNO_x during these two months is shown in Plates 1c and 1d. Because of the different contribution of CG lightning versus IC lightning to the NO_x concentrations the distribution of lightning activity (Plates 1a and 1b) is not identical to the distribution of LNO_x production (Plates 1c and 1d).

Using all 12 months of 1988, the annual mean meridional and zonal distributions of LNO_x are shown in Figures 4a and 4b. The meridional distribution clearly shows the three main centers of LNO_x production, namely, the Americas, Africa, and the maritime continent in Southeast Asia. The minima represent the oceanic regions where little lightning is observed. The annual mean latitudinal profile for 1988 shows the maximum concentration of LNO_x in the tropics, with approximately two thirds of the total LNO_x originating between 30°S and 30°N. The two secondary maxima at approximately 50°S and 50°N result from midlatitude thunderstorms that can occur during the winter and summer seasons. As a result of larger areas of land masses in the northern hemisphere, 64% of the lightning activity (annual mean), and hence LNO_x production, occurs in the northern hemisphere. We note that a significant fraction of LNO_x, 23%, occurs in northern midlatitudes (30°N–60°N) as compared to only 6% in southern midlatitudes.

The vertical distribution of LNO_x following its production, and the immediate redistribution within the storm, is of extreme importance when considering the production of tropospheric ozone. The parameterizations used in this study only supply information on total LNO_x per grid box, although it is possible to assign part of the total LNO_x to CG flashes and part to IC flashes. However, because of the strong updrafts and downdrafts in thunderstorms it is not important at what altitude the LNO_x is introduced into the atmosphere but rather the altitude to which it is transported as a result of the redistribution of air parcels within the storm. We note that a C-shaped profile with two maxima, one at the surface and the other at the upper outflow region of the storm, was obtained from a detailed cloud model with only CG lightning sources [Lyons *et al.*, 1994].

For January and July 1988 the global monthly mean LNO_x production rates are

$$G(\text{Jan}) = (19 + 0.1(52))(6.7 \times 10^9)(2.678 \times 10^6) \cdot (10 \times 10^{16})(2.33 \times 10^{-23}) = 1.02 \text{ Tg N/month} \quad (16)$$

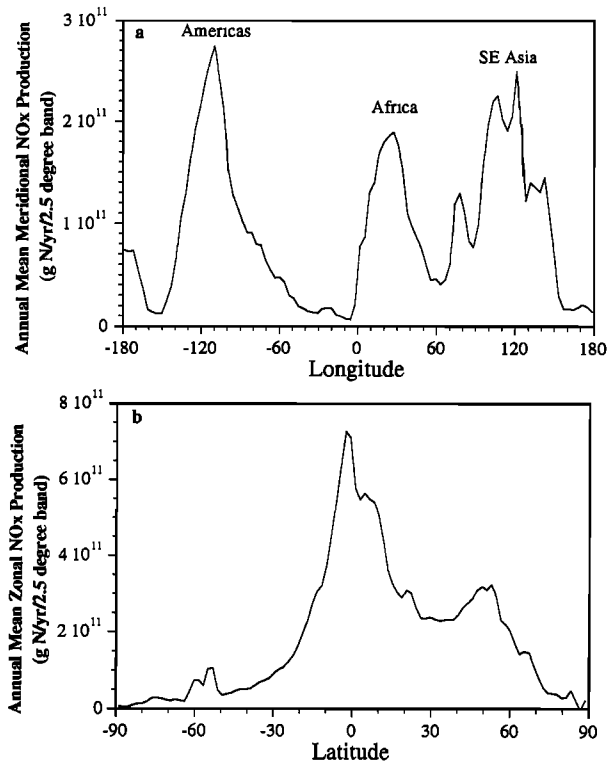


Figure 4. (a) Annual mean meridional distribution of LNO_x for 1988 and (b) annual mean zonal distribution of LNO_x for 1988.

$$G(\text{Jul}) = (30 + 0.1(71))(6.7 \times 10^9)(2.678 \times 10^6) \cdot (10 \times 10^{16})(2.33 \times 10^{-23}) = 1.55 \text{ Tg N/month} \quad (17)$$

These values represent an equivalent annual production rate of 12 Tg N/yr (Jan) and 18 Tg N/yr (Jul). The daily fluctuations in globally integrated lightning activity during these two months, together with daily fluctuation in global LNO_x production, are presented in Figure 5. Although the NO_x concentrations on a daily basis generally follow the lightning frequencies, there are periods when the trends are very different. This results from the different contribution of CG flashes and IC flashes to the total NO_x concentrations. The daily production of LNO_x appears to be episodic, with enhanced LNO_x production occurring over periods of a few days.

The above calculation of NO_x production has been done for the eight years of ISCCP data (1983–1991) (Figure 6). There appears to be a clear annual cycle in NO_x production with maximum values during the northern hemisphere summer. The production rate varies from 9 to 19 Tg N/yr, indicating a reasonable amount of variability on the annual timescale. However, the interannual variability presented in Table 1 shows much less variability. The mean annual production over 8 years is 12.2 Tg N/yr and ranges from 11.3 to 13.1 Tg N/yr.

6. Discussion

Although our calculation of global LNO_x production appears to agree with a large range of previous studies, we feel this is purely fortuitous. Previous studies have made two incorrect assumptions that appear to negate each other. First, the energy per flash in previous studies has been underesti-

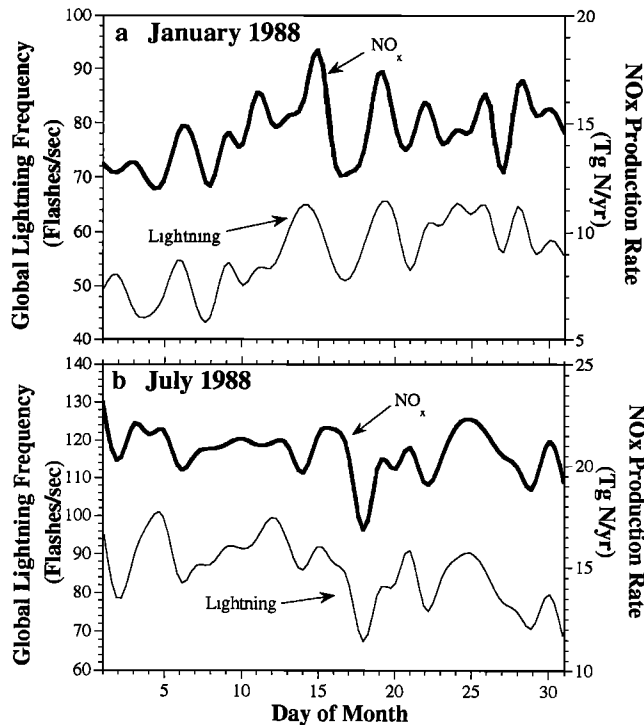


Figure 5. Daily fluctuations in global lightning frequencies and global LNO_x production for (a) January 1988 and (b) July 1988.

ated, possibly due to the fact that often the total energy in a lightning flash is derived from the optical energy that represents less than 1% of the total energy. In this paper we used observations of lightning from the U.S. Lightning Detection Network [Orville, 1991a], together with an empirical model of a lightning discharge to determine the mean energy in typical lightning flashes. We find that although the total energy can range from 10^9 to 10^{10} J [Uman, 1987], an acceptable mean value for a cloud-to-ground flash is 6.7×10^9 J. This mean value resulted from an observed mean peak current of 35.7 kA and a mean breakdown potential of 3×10^8 V. The mean peak current is obtained from lightning observations in the United States and may be different on a global scale. Furthermore, the peak currents and multiplicity of the flashes are obtained from the National Lightning Detection Network that may be biased to high-intensity flashes, while missing the smaller less intense strokes. Other studies have found mean peak currents ranging from 20 to 30 kA, although these studies observed individual

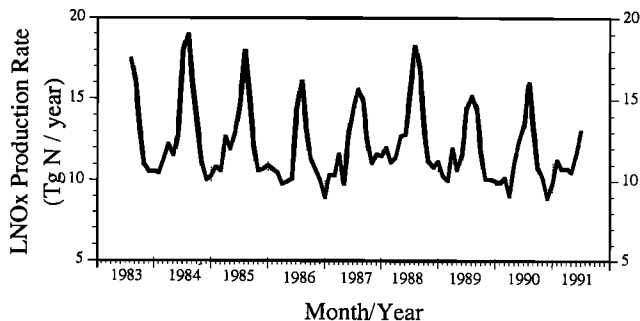


Figure 6. Monthly fluctuations in global LNO_x production from July 1983 to June 1991.

storms at single locations. The breakdown potential was determined from observations of the maximum electric field in clouds, multiplied by the altitude of the negative charge center in thunderstorms. Calculating the potential in this way is only correct when the radius of the negative charge region in the cloud base is larger than the height of this charged region above the ground. Whether these observations and assumptions are consistent on a global scale is difficult to say. Second, the global frequency of NO_x-producing flashes has been overestimated in previous studies. Although there appears to be of the order of 100 flashes per second around the globe, 70% of these flashes are intracloud discharges, with possibly only 10% of the NO_x-producing power of CG flashes. These two negating effects appear to have resulted in previous studies arriving at global estimates of LNO_x production similar to the values obtained in this study.

The global distribution of lightning was approximated using observed cloud data from the International Satellite Cloud Climatology Project (ISCCP) together with a lightning parameterization derived by Price and Rind [1992a, 1993]. The global lightning frequencies are calculated over an 8-year period (1983–1991) and provide lightning data at spatial scales from regional to global and at temporal resolutions of daily to interannual. Although the global calculations of lightning frequencies using the ISCCP data have been shown to be in good agreement with observations, there are problems with the available observations of global lightning used for verification. The detection efficiencies of the satellite-based sensors are uncertain [Turman and Edgar, 1982], and the observations are made at only a few specific local times during the day [Orville and Henderson, 1986]. Furthermore, no true climatology is available since the observations only cover a few isolated years. It is therefore difficult to know with certainty how well we are simulating global lightning distributions.

The relationship used to derive the fraction of CG lightning in a thunderstorm may need refining in the future as more lightning data become available. All the relationships used in the global simulations of lightning presented in this paper were derived using lightning data mainly from the United States. These relationships may be different in other regions of the globe. In particular, because of the difference in energies between CG and IC flashes, knowing the fraction of global lightning that is intracloud is very important for the NO_x production. It is uncertain how much energy exists in an IC flash when compared with a CG flash. We have assumed a ratio of 1/10; however, this ratio may change as our understanding of intracloud lightning improves.

We believe that the largest uncertainty in this study is the value for the amount of NO produced per joule of energy. Is the production of NO_x linearly related to the energy? Would inclusion of water vapor in the theoretical and laboratory measurements result in significant enhancements of NO production? Is corona discharge a significant source of NO in thunderstorms? We used a value of 10×10^{16} molecules NO/J in all

Table 1. Annual Production of LNO_x for Years 1984 to 1990

Year	1984	1985	1986	1987	1988	1989	1990	Mean
NO _x Production, Tg N/yr	13.0	12.6	11.3	12.2	13.1	11.7	11.3	12.2

our calculations. Our range of uncertainty for this value is $\pm 50\%$, or $10 \pm 5 \times 10^{16}$ molecules NO/J. Using a value of 5×10^{16} molecules NO/J and the lowest annual production rate (1986) would imply an annual mean production rate of 5.7 Tg N/yr, while a value of 15×10^{16} molecules NO/J and the maximum annual production rate (1988) would give an annual production rate of 19.7 Tg N/yr. This would imply a probable range of 5–20 Tg N/yr.

7. Conclusions and Implications for Tropospheric Ozone

Using our best estimate of the amount of NO_x produced per unit energy (10×10^{16} molecules NO/J), we have generated regional and global NO_x distributions for all temporal scales. We have found that the annual mean production rate of NO_x by lightning (1983–1991) is between 11.3 and 13.1 Tg N/yr, with a mean of 12.2 Tg N/yr. The production of LNO_x tends to be concentrated over the tropical landmasses, with maxima over South America, Africa, and Southeast Asia. Approximately two thirds of the global LNO_x occurs in the tropics between 30°N–30°S, while the northern hemisphere produces twice as much LNO_x than the southern hemisphere. The northern hemisphere midlatitudes produces 23% of the global LNO_x, compared with 6% produced in southern hemisphere midlatitudes. This large, factor of 4, difference in lightning NO_x implies that even prior to anthropogenic influences on the Earth's climate, there existed a natural imbalance in NO_x and hence tropospheric O₃ between the hemispheres.

The efficiency of NO_x in producing O₃ in the free troposphere is large. Values of the ratio of O₃ molecules produced per NO_x emitted range from about 30 to 60 [Lin et al., 1988; Jacob et al., 1993]. A modest estimate would be that at least 50% of LNO_x is transported out of the boundary layer [Lyons et al., 1994], and thus the global mean source of free tropospheric NO_x is at least 1.7×10^9 molecules/cm²/s. The expected source of O₃ from LNO_x is then $5\text{--}10 \times 10^{10}$ molecules/cm²/s, which is larger than the stratospheric source of about $4\text{--}6 \times 10^{10}$ molecules/cm²/s derived from either models [Stordal et al., 1995] or data analysis [Murphy and Fahey, 1994]. This northern hemisphere source of O₃ would be a third larger than the average (twice that in the southern hemisphere) and clearly dominate the stratospheric source. In the preindustrial atmosphere, lightning is expected to be the dominant source of NO_x and hence in situ O₃ production. We would thus predict that tropospheric ozone would have on the average a ratio of 3:2 between hemispheres, even in the absence of human activities over the past century.

An annual climatology of global lightning and LNO_x production has been generated using the above methodology and is now freely available to interested parties through the Global Emissions Inventory Activity (GEIA) database or from the authors. The above global results compare favorably with part 2 of this study [Price et al., this issue] where an alternative method is presented to obtain global LNO_x production rates using the global atmospheric electric circuit. A modeling study looking at the impact of these results on NO_x abundances and O₃ production will be presented in part 3 of this study.

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