JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 106, NO. D21, PAGES 27,701-27,710, NOVEMBER 16, 2001

NO_x production by lightning over the continental United States

Donald W. Bond,[1] Renyi Zhang,[1] Xuexi Tie,[2] Guy Brasseur,[2,3] Gary Huffines,[4] Richard E. Orville,[1] and Dennis J. Boccippio[5]

Abstract. The production of NO_x by lightning over the contiguous United States has been evaluated by using combined ground-based and satellite lightning measurements. The lightning data from the National Lightning Detection Network (NLDN) over the period of 1995-1999, along with a ratio of intracloud (IC) to cloud-to-ground (CG) flashes derived in conjunction with satellite lightning measurements from the Optical Transient Detector (OTD), are analyzed to obtain the number of CG and IC flashes. The average annual lightning counts over the 5-year period are about 23 million for CG flashes and 55 million for IC flashes. The resulting lightning distributions are employed to calculate the production of NO_x, assuming a NO production rate of 6.7×10^{26} molecules for each CG flash and 6.7×10^{25} molecules for each IC flash. NO_x production by lightning varies seasonally in accordance with the lightning distribution, with the maximum production occurring in the summer (June, July, and August) and in the Southeast. CG flashes produce more NO_x than IC flashes despite fewer CG flashes by a factor of 2 or more. The geographical and seasonal production of NO_x by lightning is compared to NO_x emissions from other sources (i.e., from anthropogenic, soil, and biomass-burning emissions). The results indicate that regional emissions of NO_x by lightning can be significant in the summertime and may play a critical role in ozone formation in the free troposphere. Our estimate of NO_x emission by lightning over the United States would decrease by an order of magnitude if we use the production rates from a recent laboratory study [*Wang et al.*, 1998] that are significantly lower than previous estimates.

1. Introduction

The atmosphere serves as an oxidizing medium in the global biogeochemical cycles [*Intergovernmental Panel on Climate Change* (IPCC), 1996; *Schlesinger*, 1997]. Reduced chemical species emitted by the biosphere and by human activities are oxidized in the atmosphere. The oxidizing capacity of the atmosphere is related to the abundance of the oxidants (such as reactive nitrogen oxides $NO_x = NO + NO_2$), which is determined by their production and removal processes. In particular, reactive nitrogen oxides play an important role in influencing the ozone concentration [*Crutzen*, 1970]. In the troposphere and stratosphere, NO_x is closely related to the ozone chemistry via two separate processes. In regions of high NO_x concentrations, ozone is produced photochemically in the cycling of NO to NO_2, which is facilitated by peroxy radicals formed during oxidation of carbon monoxide, methane, and other volatile organic compounds (VOCs), while in regions of low NO_x concentrations, ozone is catalytically destroyed. Nitrogen oxides are also intricately linked to the hydroxyl radical OH, another key atmospheric oxidizing species. The reaction between NO_2 and OH leads to the formation of relatively stable nitric acid HNO_3, which can be removed from the atmosphere by precipitation and hence provides an important fixed nitrogen for the biosphere. Also, since O_3 strongly absorbs the Earth's infrared radiation, knowledge of the regional and global NO_x distribution is important for climate studies.

Nitrogen oxides are emitted into the atmosphere from natural and anthropogenic sources, that is, from fossil fuel combustion, biomass burning, oxidation of atmospheric ammonia, and lightning [*Seinfeld and Pandis*, 1998]. In addition, transport of NO_x from the stratosphere and aircraft emissions could also be important sources in the upper troposphere [e.g., *Brasseur et al.*, 1996; *Strand and Hov*, 1996; *Penner et al.*, 1999]. Previous studies have indicated that lightning can lead to a significant enhancement of NO_x in the middle and upper troposphere [*Ridley et al.*, 1996; *Huntrieser et al.*, 1998; *Pickering et al.*, 1998; *Zhang et al.*, 2000]. To obtain an estimate of the production of NO_x by lightning on a regional or global scale, several parameters need to be assessed and defined, including the production rate of NO_x per joule of energy, the average energy dissipated per lightning flash, and the lightning distribution. NO_x is thought to be produced primarily in the completely dissociated channel of a lightning discharge through the recombination reactions of atomic oxygen and nitrogen [*Zel'dovich and*

[1]Department of Atmospheric Sciences, Texas A&M university, College Station, Texas, USA.
[2]National Center for Atmospheric Research, Boulder, Colorado, USA.
[3]Max-Planck Institute of Meteorology, Hamburg, Germany.
[4]Department of Engineering Physics, Air Force Institute of Technology, Wright-Patterson Air Force Base, Ohio, USA.
[5]Global Hydrology and Climate Center, NASA-Marshall Space Flight Center, Huntsville, Alabama, USA.

Paper number 2000JD000191.
0148-0227/01/2000JD000191$09 00

Raizer, 1966]. The process involves "freezing out" a few percent of NO_x as the temperature of the lightning channel decreases to about 3000 K. At that point in time, the volume of hot gas produced, which is related to the amount of energy deposited on the gas by the lightning stroke, controls the amount of NO produced per joule of energy input. Other ambient factors may also influence the amount of NO_x produced per flash, including pressure, relative humidity, and the duration of the discharge. In addition, NO_x can also be produced by a mechanism involving ion-molecule reactions from ion and photon generation induced by lightning discharge [*Griffing*, 1977; *Boldi*, 1992].

A number of studies have investigated the production rate of NO_x by a single lightning discharge. In laboratory experiments NO and NO_2 molecules from induced electrostatic discharges have been measured and related to the energy of the discharge to yield the production rate in number of NO_x molecules per joule [*Chameides et al.*, 1977; *Levine et al.*, 1981; *Peyrous and Lapeyre*, 1982; *Stark et al.*, 1996; *Wang et al.*, 1998]. Theoretical models, on the other hand, have simulated the energy dissipation of the lightning channel either by a shock wave mechanism [*Tuck*, 1976; *Chameides et al.*, 1977; *Chameides*, 1979] or by an ohmic heating model [*Hill*, 1971; *Griffing*, 1977; *Hill et al.*, 1980]. Additional theoretical work [*Goldenbaum and Dickerson*, 1993] concluded that the NO_x production rate is strongly dependent on the initial energy intensity in the heated channel. Also, field measurements of NO_x production have been reported [*Noxon*, 1976; *Drapcho et al.*, 1983; *Franzblau and Popp*, 1989; *Cook et al.*, 2000]. When applying these results to actual lightning flashes in the atmosphere, however, there are several complicating factors. First, the characteristics of lightning such as surface-to-volume ratio, voltage-current relationship, and thermal properties in laboratory and theoretical simulations may differ from real lightning discharge and hence may affect the NO_x production rate [*Liaw et al.*, 1990; *Biazar and McNider*, 1995; *Price et al.*, 1997]. Also, intracloud (IC) flashes are thought to have significantly lower energies than cloud-to-ground (CG) discharges and hence have a lower NO_x production efficiency [*Borucki and Chameides*, 1984]; it has been suggested that the amplitude of the radiation field for a typical CG flash in the 1-10 kHz range is much larger than that of an IC flash [*Uman*, 1987]. Furthermore, there is an uncertainty concerning NO_x production by the corona sheath surrounding the lightning channel [*Boldi*, 1992].

Two earlier studies have assessed the importance of lightning NO_x production on regional NO_x budgets. The work of *Pierce and Novak* [1991] used the National Lightning Detection Network (NLDN) data for 2 weeks in the Northeast to estimate NO_x emissions in regional air quality models. *Biazar and McNider* [1995] reported regional estimates of lightning production of NO_x by using data from the NLDN for June, July, and August from 1989 to 1992 and suggested that lightning can be a nonnegligible NO_x source in the summertime over the southeastern portion of the United States. In those previous studies, however, it was assumed that all flashes are of the CG type, with no IG contribution included.

In this study we estimate the geographical and seasonal distributions of NO_x production by lightning over the contiguous United States by using combined ground-based and satellite lightning measurements. The NLDN lightning data, along with an IC/CG ratio derived in conjunction with satellite lightning measurements, are analyzed. The resulting lightning distributions are then used to estimate the production of NO_x by lightning. A comparison of the NO_x production by lightning is made with the other major sources of nitrogen oxides to assess the importance of lightning as a natural NO_x source and its role in tropospheric ozone formation.

2. Data and Method

This study uses monthly lightning data from the National Lightning Detection Network (NLDN) from 1995 to 1999, along with an IC/CG ratio derived in conjunction with the satellite data from the Optical Transient Detector (OTD), to obtain the geographical and seasonal distributions of CG and IC flashes. The NLDN is operated by the GeoMet Data Services, Inc. (now Global Atmospherics, Inc.), Tucson, Arizona. The network consists of over 100 wideband magnetic direction finders [*Orville*, 1994], augmented by time-of-arrival (TOA) sensors beginning in July 1994. The direction finders consist of two orthogonal magnetic loop antennas, a flat plate antenna, and associated electronics to process the incoming signal. Within a range of 600 km, the location and polarities of the incoming lightning waves are determined. Locations of the magnetic direction finders in the contiguous United States have been published previously [*Orville*, 1994]. The NLDN continuously monitors cloud-to-ground flash occurrence with little spatial bias. The principles of the magnetic direction finder, detection efficiency, and location errors have been discussed previously [e.g., *Krider et al.*, 1976, *Orville*, 1994]. Location errors are typically assumed to be of the order of 10 km [*Pyle*, 1995; *Orville and Silver*, 1997]. In the present analysis, we assume that a 85% detection efficiency applies throughout the network, consistent with recent measurements by *Cummins et al.* [1998]. All flash densities were multiplied by a factor of 1.18 to correct for the detection efficiency. It is likely that the detection efficiency may be different for negative and positive flashes, as previously suggested by *Hojo et al.* [1989]. However, in the absence of separate measurements of negative and positive detection efficiencies for the advanced lightning direction finders (ALDF), we assume that the detection efficiency is the same for both types of flashes.

We use an IC/CG ratio derived in conjunction with satellite lightning measurements from the Optical Transient Detector (OTD) [*Boccippio et al.*, 2001] to obtain the number of IC flashes. OTD is the first long-term observer of lightning from space on the MicroLab 1 satellite [*Goodman et al.*, 1988; *Christian et al.*, 1989]. It detects both IC and CG flashes by observing momentary changes in the neutral oxygen emission line at 777.4 nm that indicates the occurrence of lightning [*Christian et al.*, 1989]. The satellite was launched on April 3, 1995 into an orbital altitude of 710 km and an inclination of 70° [*Christian and Latham*, 1998]. The satellite orbits the Earth approximately once every 100 min. The OTD sensor has a 100° viewing angle that facilitates a 1300 × 1300 km viewing footprint. This allows viewing of approximately 1/300 of the total surface area of the Earth at a particular moment. The instrument has a spatial resolution of 10 km in the horizontal and a temporal resolution of 2 ms. The OTD

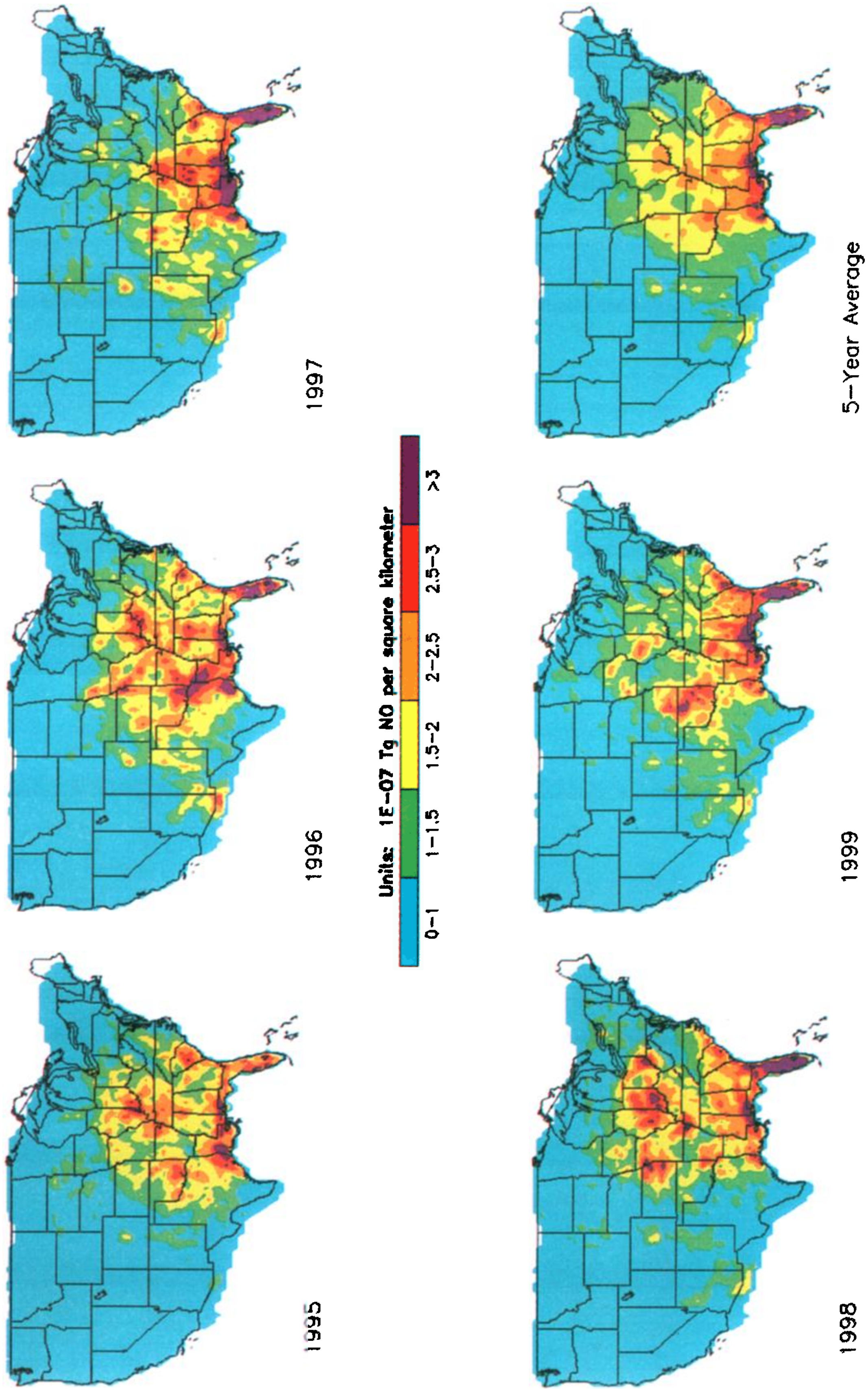

Plate 1. Geographical distributions of NO_x production by lightning over the contiguous United States from 1995 to 1999. The values are sums of NO production per square kilometer in 10^{-7} Tg.

detects lightning flashes with an efficiency between 40% and 65%, depending on viewing conditions such as Sun glint and radiation [*Boccippio et al.*, 2001]. Combining 4 years of OTD and NLDN lightning data, *Boccippio et al.* [2001] obtained the geographical distribution of the climatological IC/CG ratio over the continental United States. The average value of the IC/CG ratio ranges from 2.64 to 2.94, with a standard deviation of 1.1-1.3 and anomalies as low as 1.0 or less over the Rocky and Appalachian Mountains and as high as 8-9 in the central and upper Great Plains. In this study, both the data for the NLDN CG flashes and the IC/CG ratios from *Boccippio et al.* [2001] were composited into a 0.5° x 0.5° grid, and the product of those two parameters yields the number of IC flashes.

We have considered available results from laboratory experiments, field measurements, and modeling simulations, in order to estimate the NO_x production by lightning flashes. *Price et al.* [1997] suggested that the best representative values of NO_x production are 6.7 x 10^{26} and 6.7 x 10^{25} NO molecules for each CG and IC flash, respectively, on the basis of available laboratory experiments, theoretical simulation, and field measurements of NO production by naturally occurring or artificially triggered sparks. Those values are consistent with recent field measurements [*Liaw et al.*, 1999]. In addition, using the same values for the NO production rates by IC and CG flashes, *Nesbitt et al.* [2000] obtained a global lightning NO_x emission of 8.8 Tg N yr^{-1}, based on an analysis of the OTD lightning data. The latter lightning NO_x emission results have been incorporated into a global atmospheric chemical transport model (CTM), and the model simulations of the NO_x concentrations are in good agreement with aircraft NO_x measurements over several continents [*Tie et al.*, 2001]. Hence we have adopted the production rates suggested by *Price et al.* [1997] to calculate NO_x production by IC and CG flashes in this study.

In order to establish the importance of lightning as a natural source for NO emission over the contiguous United States, comparisons were made between NO_x production by lightning and by other sources, including anthropogenic, soil, and biomass-burning emissions. The NO_x emission data from anthropogenic activity, soil release, and biomass burning are taken from the recent compilations by *Olivier et al.* [1996, 1999], which have been improved over the 1985 modeler's inventory of the National Acid Precipitation Assessment Program (NAPAP) and the 1985 Global Emission Inventory Activity (GEIA).

Table 1. The Average Monthly Number of Lightning Flashes in the Period 1995-1999 over the Contiguous United States

Month	Cloud-to-Ground	Intracloud	Total
Jan.	295,673	768,087	1,063,760
Feb.	202,776	507,802	710,578
March	507,072	1,274,444	1,781,516
April	998,477	2,516,905	3,515,382
May	2,684,226	6,648,858	9,933,084
June	4,484,193	10,773,539	15,257,732
July	5,884,319	13,791,942	19,676,261
Aug.	4,557,351	11,058,546	15,615,897
Sept.	1,980,822	4,989,793	6,970,615
Oct.	577,878	1,517,973	2,095,851
Nov.	194,102	530,066	724,168
Dec.	114,714	324,219	438,933
Annual	22,481,603	54,702,174	77,183,777

3. Results and Discussion

3.1. IC and CG Flashes

Table 1 summarizes the 5-year average seasonal variation of lightning flashes and the contributions from IC and CG flashes to the total lightning over the contiguous United States. The CG flashes are obtained from the NLDN data, and IC flashes are derived from the NLDN data along with the IC/CG ratio reported by *Boccippio et al.* [2001]. The results presented in Table 1 have been corrected for detection efficiency of the NLDN data. On average, at least 15 million flashes occur over the United States each month during the summer, reaching almost 20 million flashes in July. The high lightning activity in June, July, and August is consistent with the notion that more lightning occurs in the summertime due to more deep convection, which accelerates charge separation by interacting cloud particles in thunderstorms [*Williams et al.*, 1991].

There is a slight interannual variability in the lightning activity. Over the 5-year period we estimate that there are on average 23 million CG flashes and 55 million IC flashes, with standard deviations of 1.3 million for CG flashes and 2.7 million for IC flashes. The 5-year average IC/CG ratio is about 2.4, that is, an average of 2.4 IC flashes for each CG flash. The highest lightning count during this period occurs in 1998 with 24 million CG flashes and 60 million IC flashes, and the lowest lightning count takes place in 1995 with 21 million CG flashes and 50 million IC flashes. The average number of CG flashes of 23 million obtained from 1995 to 1999 is close to the number previously published by *Orville and Silver* [1997], who reported an average of about 22 million CG flashes from 1992 to 1995.

3.2. Climatology of Lightning NO_x Production

Plate 1 depicts the geographical distribution of NO_x production by lightning from 1995 to 1999. The climatology of NO_x production by lightning was obtained by assuming the production rates of 6.7 x 10^{26} and 6.7 x 10^{25} NO molecules for each CG and IC flash, respectively. The plotted values are sums of the NO production per square kilometer. It is apparent in this plate that the dominant lightning NO_x production area is in the Southeast. On the other hand, annual NO_x emissions by lightning are relatively small over the majority of the western, Great Plains, and northeastern regions.

It is also seen from Plate 1 that there is a slight interannual variability in the distribution of NO_x production by lightning between 1995 and 1999. For example, the NO_x emission by lightning in 1997 is noticeably weak in parts of the midwestern region (i.e., Missouri, Iowa, Illinois, etc.) compared to the other four years, but is apparently the

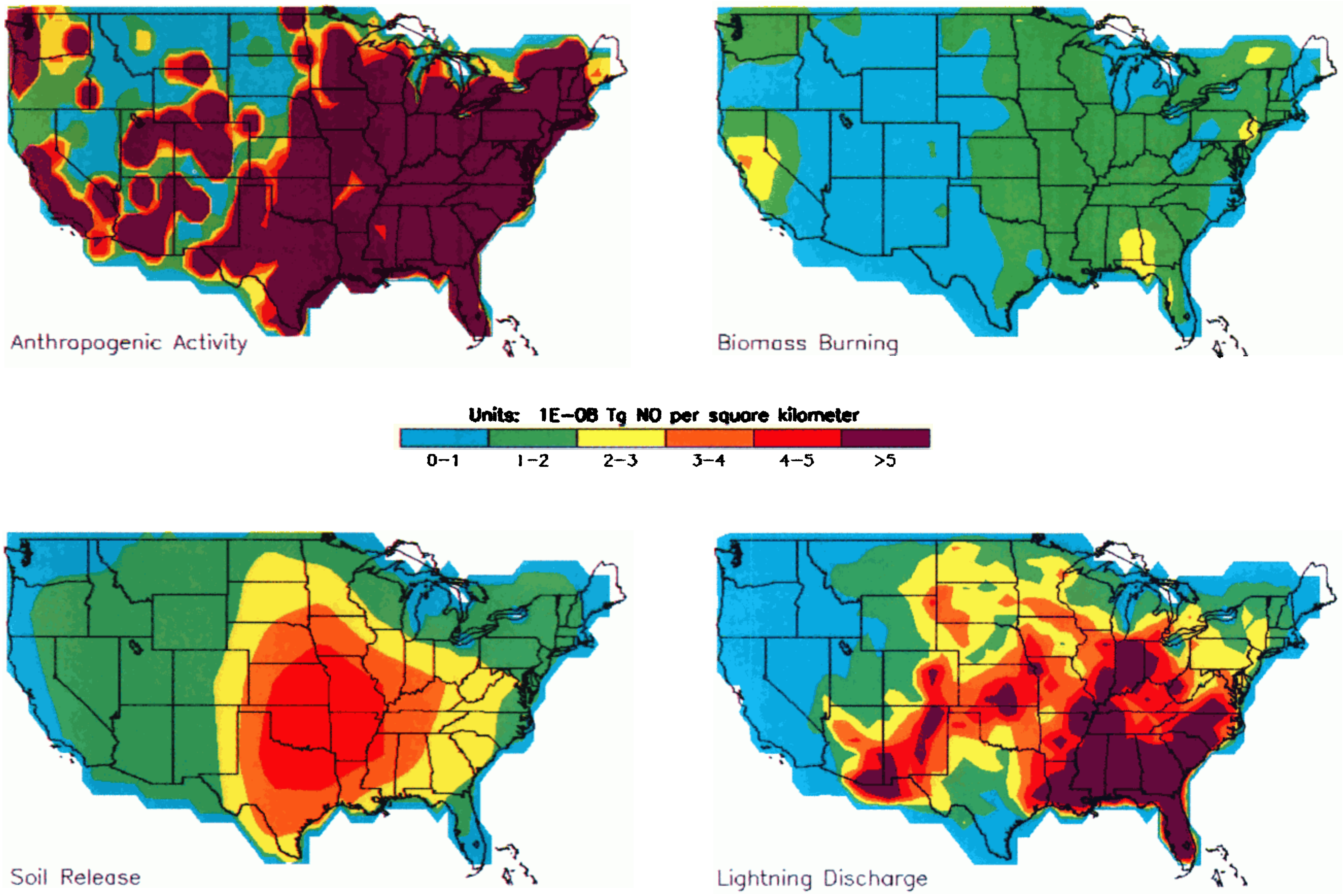

Plate 2. Geographical distributions of NO_x emissions from the various sources over the United States in July. The values are sums of NO production per square kilometer in 10^{-8} Tg.

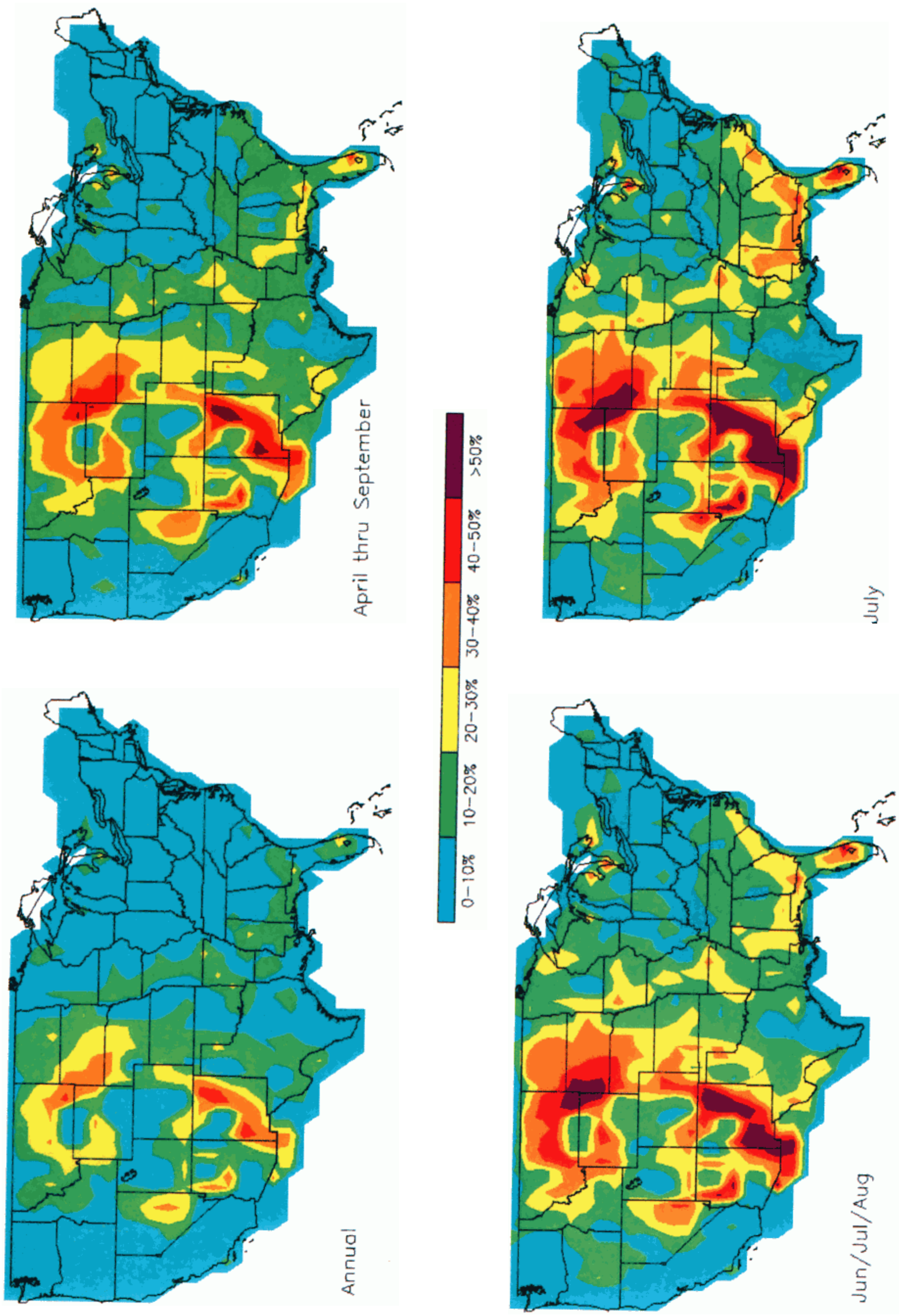

Plate 3. Contribution of NO_x production by lightning to the total NO_x budget over different time periods for the United States. The percentage is computed from the ratio of lightning-produced NO_x to that produced by the combined four sources (i.e., from anthropogenic, soil, biomass-burning, and lightning emissions).

Table 2. Nitrogen Oxide Emissions by Sources[a]

Month	Lightning	Biomass Burning	Soil Release	Anthropogenic Activity
Jan.	0.012	0.045	0.017	1.422
Feb.	0.008	0.041	0.019	1.279
March	0.021	0.046	0.030	1.390
April	0.041	0.048	0.053	1.306
May	0.111	0.054	0.089	1.308
June	0.183	0.067	0.145	1.229
July	0.240	0.088	0.193	1.249
Aug.	0.187	0.096	0.181	1.253
Sept.	0.082	0.098	0.114	1.242
Oct.	0.024	0.100	0.065	1.325
Nov.	0.008	0.068	0.029	1.326
Dec.	0.005	0.050	0.019	1.405
Annual	0.922	0.801	0.953	15.733

[a] Units are teragrams NO.

strongest in the Southeast over the 5-year period. Nevertheless, the interannual variability in the NO_x emission is small compared to the seasonal variation of the lightning NO_x emission (see also Table 2).

It is interesting to note in Plate 1 that on the 5-year average the geographical area centered on Houston, Texas, shows an anomalously high NO_x production. Also the geographical area located near Denver, Colorado, corresponds to an anomalously high NO_x production. The enhanced lightning activity in Houston has been explained jointly due to the enhanced convergence associated with the urban heat island effect and high levels of air pollutants from anthropogenic sources [*Orville et al.*, 2001].

Table 2 presents the seasonal variation of NO_x production by lightning. The production listed in Table 2 is the average value over the 5-year period. NO_x production by lightning exhibits a strong seasonal variation, consistent with the seasonal variation of lightning activity (Table 1). The annual production by lightning is 0.92 Tg NO yr^{-1}, with 0.18 and 0.74 Tg NO yr^{-1} from IC and CG flashes, respectively. The production is maximal in July, equivalent to 26% of the annual lightning production.

3.3. Lightning Contribution to the NO_x Budget

Table 2 also includes the seasonal variations of NO_x emissions from the other sources. The data of NO_x emissions from anthropogenic activity, soil release, and biomass burning are taken from the recent compilations by *Olivier et al.* [1996, 1999]. NO_x emissions from biomass burning and soil release both show a seasonal variation. For biomass burning, the production reaches the highest levels in September and October, whereas the soil release peaks in July and August. On the other hand, the anthropogenic emission dominates throughout the year, with little seasonal variation. The production of NO_x by lightning is slightly higher than that from soil release in the summer months (i.e., June, July, and August), although the annual contributions from those two sources are nearly comparable (0.92 Tg NO yr^{-1} for lightning and 0.95 Tg NO yr^{-1} for soil release). The annual emissions from both lightning and soil are slightly higher than that from biomass burning (0.80 Tg NO yr^{-1}). The anthropogenic emissions of NO_x are much stronger than the other three sources, with an estimated annual emission of about 16 Tg NO yr^{-1}. The lightning production is only 6% of the annual anthropogenic emission, but is as high as 20% of the anthropogenic emission in July.

Plate 2 compares the geographical distributions of NO_x emissions from the various sources in July. It is seen from the plate that the anthropogenic emission dominates over the majority of the contiguous United States, except in parts of the Great Plains and the western region. Lightning emission is also intensive in the most of the Southeast and some portions of the midwestern region. In addition, the mountain areas of Arizona and New Mexico exhibit significant NO_x production in July. NO_x production from soil release in the summertime is located broadly in the midcentral region, but is less intensive compared to the emissions from anthropogenic activity and lightning. The contribution of NO_x production from biomass burning is much smaller in July, except in parts of California.

To assess the contribution of lightning to the NO_x budget, we plot the percentage of NO production by lightning to the total NO emission from the combined four sources (Plate 3). Since there is a strong annual variability in lightning, we consider the lightning contribution within several time periods. It is clear from Plate 3 that on an annual basis lightning typically contributes less than 10% of the NO_x budget in most of the eastern and western parts of the United States. Apparently, this is a result of the dominant anthropogenic emissions in those regions. There is, however, a narrow band extending from New Mexico to Montana with large lightning contribution. For example, in the Rocky Mountain area of New Mexico, the percentage of lightning production reaches as high as 50%. The large contribution of NO_x production by lightning in this region can be explained as being due to both a lesser amount of anthropogenic emission and more summertime lightning activity (Plate 2). Similar features are also evident in the plots of April to September, June to August, and July.

The contribution by lightning appears to increase substantially in the summertime (July or June to August) in the Southeast. In July the percentage of NO_x production ranges from 10% to 50% in this region. The highest values occur along the Southeast coastal regions. Apparently, those high percentages of NO_x production by lightning are attributed to the fact that the lightning activity in July is maximal in the Southeast, even when the anthropogenic source is still significant. Hence in the summertime, lightning constitutes a significant source for NO_x emission, particularly in the Southeast.

4. Discussion

The present results indicate that lightning can be a major source of NO_x emissions on a regional scale. On an annual basis, lightning NO_x production consists of 5% of the total NO_x emissions and the anthropogenic emission dominates over most portions of the contiguous United States. In the summer months, however, the lightning contribution increases substantially, reaching as high as 14% of the total NO_x emissions in July. In addition, our results indicate that lightning NO_x emission dominates in the Southeast, and hence plays a significant role in the regional emissions there. The annual production by lightning in the United States is 0.92 Tg NO yr^{-1}, with 0.18 and 0.74 Tg NO yr^{-1} from IC and CG flashes, respectively. It is also important to note that the vertical distribution of NO_x emission by lightning may have important implications on assessing the role of lightning NO_x production on tropospheric ozone formation. Since the lifetime of NO_x varies considerably with altitudes, from only a few hours near the planetary boundary layer up to a few days in the upper troposphere, NO_x emitted directly into the middle and upper troposphere will be more efficient in producing ozone [*Tie et al.*, 2001]. Lightning is the only source that results in direct emissions of NO_x into the free troposphere, whereas the other three sources emit NO_x primarily within the planetary boundary layer.

Two recent studies [*Price et al.*, 1997; *Nesbitt et al.*, 2000] have assessed the global production of NO_x by lightning discharges, which can also be used to infer the lightning NO_x production within the United States. *Price et al.* [1997] parameterized the global and seasonal distributions of lightning-produced NO_x on the basis of the distributions of electrical storms, their heights, and the physical properties of lightning strokes. *Nesbitt et al.* [2000] estimated the global NO_x production by lightning by using the satellite lightning data from the Optical Transition Detector (OTD) in 1996. A comparison of the geographical distributions of NO_x production by lightning in the United States between this study and those previously reported reveals some similarity. For example, all the studies indicate that the highest production occurs in the southeast portion of the United States. The annual NO_x production over the United States estimated from the two previous studies is 2.2 Tg NO yr^{-1} from the OTD data and 0.86 Tg NO yr^{-1} from the parameterization of *Price et al.* [1997]. Those values are somewhat different from the present estimate. Nevertheless, the use of the NLDN data significantly reduces the uncertainty in the seasonal and geographical distributions of lightning over the United States. The method to determine the lightning distributions suggested by *Price et al.* [1997] relies on simulation of the convective storms, while the use of the OTD data requires major extrapolation because of the relatively small viewing area of the OTD satellite [*Nesbitt et al.*, 2000].

Although using the NLDN lightning data significantly reduces the uncertainty associated with lightning detection over the United States, there are still uncertainties in our estimate of NO_x production made in this study, mainly associated with the use of NO production per flash [Bradshaw et al., 2000]. If the NO production values of 6.2×10^{25} and 8.7×10^{24} molecules for each CG and IC flash on the basis of the laboratory data recently published by *Wang et al.* [1998] are used, we obtain an annual NO_x production of 0.09 Tg NO yr^{-1} by lightning over the United States. This value is about an order of magnitude smaller than that estimated using the recommendation by *Price et al.* [1997]. Hence the NO production by CG and IC flashes should be better defined by future experimental, field, and theoretical studies.

5. Conclusions

We have evaluated the production of NO_x by lightning over the contiguous United States, using lightning data from the National Lightning Detection Network (NLDN) over the period of 1995-1999, along with the ratio of IC/CG flashes derived in conjunction with satellite lightning measurements. We estimate that average annual lightning counts over the 5-year period are about 23 million for CG flashes and 55 million for IC flashes. The production of NO_x by lightning varies seasonally and geographically, in accordance with the lightning activity. The highest lightning production occurs in the summer and over the southeastern part of the United States. The annual production by lightning in the United States is estimated as 0.92 Tg NO yr^{-1}, with 0.18 and 0.74 Tg NO yr^{-1} from IC and CG flashes, respectively. Our results indicate that regional emissions of NO_x by lightning can be significant in the middle and upper troposphere and hence play a critical role in ozone formation. The uncertainty of the estimate of NO_x production by lightning over United States is mainly associated with the assumed NO production rates used. Our estimate of NO_x emission by lightning over the United States would decrease by an order of magnitude if we use the production rates from a recent laboratory study [*Wang et al.*, 1998] that are significantly lower than previous estimates.

Acknowledgments. This work was supported by the NASA New Investigator Program in Earth Sciences and the Texas Air Research Center (TARC). The NLDN lightning data were obtained from Global Atmospherics, Inc., Tucson, Arizona. The Texas A&M Lightning Program was supported by NSF (ATM-9806189) and NOAA (cooperative agreement NA87WA0063). We also acknowledge the three referees for providing valuable comments on the manuscript.

References

Biazar, A.P., and R.T. McNider, Regional estimates of lightning production of nitrogen oxides, *J Geophys. Res.*, *100*, 22,861-22,874, 1995.

Boccippio, D.J., K.L. Cummins, H.J. Christian, and S.J. Goodman, Combined satellite and surface-based estimation of intracloud/cloud-to-ground lightning ratio over the continental United States, *Mon. Weather Rev.*, *129*, 108-122, 2001.

Boldi, R.A., A model of the ion chemistry of electrified convection, Ph.D. thesis, Mass. Inst. Tech., 1992.

Borucki, W.J., and W.L. Chameides, Lightning: Estimates of the rates of energy dissipation and nitrogen fixation, *Rev. Geophys.*, *22*, 363-372, 1984.

Bradshaw, J., D. Vavis, G. Grodzinsky, S. Smyth, R. Newell, S. Sandholm, and S. Liu, Observed distributions of nitrogen oxides in the remote free troposphere from the NASA global tropospheric experiment programs, *Rev. Geophys*, *38*, 61-116, 2000.

Brasseur, G.P., J.F. Muller, and C. Granier, Atmospheric impact of NO_x emission by subsonic aircraft: A three-dimensional model study, *J. Geophys. Res*, *101*, 1423-1428, 1996.

Chameides, W.L., Effect of variable energy on nitrogen fixation in instantaneous linear discharges, *Nature*, *277*, 123-125, 1979.

Chameides, W.L., D.H. Stedman, R.R. Dickerson, D.W. Rusch, and R.J. Cicerone, NO_x production in lightning, *J. Atmos Sci*, *34*, 143-149, 1977.Christian, H.J., R. J. Blakeslee, and S.J. Goodman, The detection of lightning from geostationary orbit, *J Geophys. Res*, *94*, 13,329-13,337, 1989.

Christian, H.J., and J. Latham, Satellite measurements of global lightning, *Q. J. R. Meteorol. Soc.*, *124*, 1771-1773, 1998.

Christian, H.J., R. J. Blakeslee, and S.J. Goodman, The detection of lightning from geostationary orbit, *J. Geophys Res.*, *94*, 13,329-13,337, 1989

Cook, D.R., Y.P. Liaw, D.L. Sisterson, and N.L. Miller, Production of nitrogen oxides by a large spark generator, *J. Geophys. Res*, *105*, 7103-7110, 2000.

Crutzen, P.J., The influence of nitrogen oxides on the atmospheric ozone content, *Q. J R Meteorol. Soc.*, *96*, 320-327, 1970.

Cummins, K. L., M. J. Murphy, E. A. Bardo, W. L. Hiscox, R. B. Pyle, and A. E. Pifer, A combined TOA/MDF technology upgrade of the U.S. National Lightning Detection Network, *J. Geophys. Res*, *103*, 9035-9044, 1998.

Drapcho, D.L., D.L. Sisteron, and R. Kumar, Nitrogen fixation by lightning activity in a thunderstorm, *Atmos. Environ.*, *17*, 729-734, 1983.

Franzblau, E., and R.R Popp, Nitrogen oxides produced from lightning, *J Geophys. Res*, *94*, 11,089-11,104, 1989.

Goldenbaum, G.C., and R R. Dickerson, Nitric oxides production by lightning discharges, *J Geophys Res*, *98*, 18,333-18,338, 1993.

Goodman, S. J., H.J. Christian, and W.D. Rust, A comparison of the optical pulse characteristics of intracloud and cloud-to-ground lightning as observed above the clouds, *J. Appl Meteorol* *27*, 1369-1380, 1988.

Griffing, G.W., Ozone and oxides of nitrogen production during thunderstorms, *J. Geophys. Res.*, *82*, 943-950, 1977.

Hill, R D., Channel heating in return stroke lightning, *J. Geophys. Res.*, *76*, 637-645, 1971.

Hill, R.D., R.G. Rinker, and H.D. Wilson, Atmospheric nitrogen fixation by lightning, *J. Atmos. Sci*, *37*, 179-192, 1980.

Hojo, J., M. Ishii, T. Kawamura, F. Suzuki, H. Komuro, and M. Shiogama, Seasonal variation of cloud-to-ground lightning flash characteristics in the coastal area of the Sea of Japan, *J. Geophys. Res.*, *94*, 13,207-13,212, 1989.

Huntrieser, H., H. Schlager, C. Feigl, and H. Holler, Transport and production of NO_x in electrified thunderstorms: Survey of previous studies and new observations at midlatitudes, *J. Geophys. Res*, *103*, 28,247-28,264, 1998.

Intergovernmental Panel on Climate Change (IPCC), *Climate Change 1995: The Science of Climate Change*, Cambridge Univ. Press, New York, 1996.

Krider, E.P., R.C. Noggle, and M.A. Uman, A gated wideband magnetic direction finder for lightning return strokes, *J. Appl. Meteorol.*, *15*, 301-306, 1976.

Levine, J.S., R.S. Rogowski, G.L. Gregory, W.E. Howell, and J. Fishman, Simultaneous measurements of NO_x, NO, and O_3 production in a laboratory discharge: Atmospheric implications, *Geophys. Res. Lett.*, *8*, 357-360, 1981.

Liaw, Y.P., D.L. Sisterson, and N.L. Miller, Comparison of field, laboratory, and theoretical estimates of global nitrogen fixation by lightning, *J. Geophys. Res.*, *95*, 22,489-22,494, 1990.

Liaw, Y.P., D.R. Cook, D.L. Sisterson, and N.L. Miller, Direct field measurements of NO_x production by lightning, *Eos, Trans. AGU*, *80(46)*, Fall Meet. Suppl., F195, 1999.

Nesbitt, S.W., R. Zhang, and R.E. Orville, Seasonal and global NO_x production by lighting estimated from the optical transient detector (OTD), *Tellus, Ser. B*, *52*, 1206-1215, 2000.

Noxon, J.F., Atmospheric nitrogen fixation by lightning, *Geophys Res Lett.*, *3*, 463-465, 1976.

Olivier, J.G.J., A.F. Bouwman, C.W.M. Van der Maas, J.J.M. Berdowski, C. Veldt, J.P.J. Bloos, A.J.H. Visschedijk, P.Y.J. Zandveld, and J.L Haverlag, Description of EDGAR version 2.0: A set of global emission inventories of greenhouse gases and ozone-depleting substances for all anthropogenic and most natural sources on a per country basis and on a 1x1 degree grid, *RIVM Rep. 771060 002/TNO-MEP Rep R96/119*, Natl. Inst. of Publ Health and the Environ., Bilthoven, Netherlands, 1996.

Olivier, J.G.J, A.F. Bouwman, J.J.M. Berdowski, C. Veldt, J.P.J. Bloos, A.J.H. Visschedijk, C.W.M. van der Maas, and P.Y.J. Zandveld, Sectoral emission inventories of greenhouse gases for 1990 on a per country basis as well as on 1x1 degree, *Environ Sci Policy*, *2*, 241-264, 1999.

Orville, R.E., Cloud-to ground lightning flash characteristics in the contiguous United States: 1989-1991, *J. Geophys Res.*, *99*, 10,833-10,841, 1994.

Orville, R E., and A.C. Silver, Lightning ground flash density in the contiguous United States: 1992-95, *Mon. Weather Rev*, *125*, 631-638, 1997.

Orville, R.E., G. Huffines, J. Nielsen-Gammon, R. Zhang, S. Allen, and W. Read, Enhancement of cloud-to-ground lightning within a coastal urban environment, *Geophys. Res. Lett.*, *28*, 2597-2600, 2001.

Penner, J. E., D. H. Lister, D.J. Griggs, D.J. Dokken, and M. McFarland, *Aviation and the Global Atmosphere A Special Report of IPCC Working Groups I and III*, Cambridge Univ. Press, New York, 1999.

Peyrous, R., and R.M. Lapeyre, Gaseous products created by electrical discharges in the atmosphere and condensation nuclei resulting from gaseous phase reactions, *Atmos Environ*, *16*, 959-968, 1982.

Pickering, K.E., Y.S. Wang, W.K. Tao, C. Price, and J.F. Muller, Vertical distributions of lightning NO_x for use in regional and global chemical transport model, *J Geophys. Res.*, *103*, 31,203-31,216, 1998.

Pierce, T.E., and J.H. Novak, Estimating natural emissions for EPA's Regional Oxidant Model, in *EPA/AWMA International Specialty Conference on Emission, Inventory Issues in the 1990s*, Environ. Prot. Agency, Washington, D.C., 1991

Price, C., J. Penner, and M. Prather, NO_x from lightning. 1. Global distribution based on lightning physics, *J. Geophys. Res*, *102*, 5929-5941, 1997.

Pyle, R, National network status, paper presented at *International Lightning Detection Conference*, Lightning Location and Prot., Inc., Tucson, Az., 1995.

Ridley, B.A., J.E. Dye, J.G. Ealega, J. Zheng, F.E. Grahek, and W. Rison, On the production of active nitrogen by thunderstorms over New Mexico, *J Geophys. Res.*, *101*, 20,985-21,005, 1996.

Schlesinger, W.H., *Biogeochemistry*, Academic, San Diego, Calif., 1997.

Seinfeld, J.H., and S.N. Pandis, *Atmospheric Chemistry and Physics*, John Wiley, New York, 1998.

Stark, M.S., J.T. Harrison, and C. Anastasi, Formation of nitrogen oxides by electrical discharges and implications for atmospheric lightning, *J. Geophys Res.*, *101*, 6963-6969, 1996.

Strand, A., and O. Hov, The impact of man-made and natural NO_x emissions on the upper tropospheric ozone: A two-dimensional model study, *Atmos. Environ.*, *30*, 1291-1303, 1996.

Tie, X.X., R. Zhang, G. Brasseur, L. Emmons, and W. Lei, Effects of lightning on reactive nitrogen and nitrogen reservoir species in the troposphere, *J. Geophys. Res.*, *106*, 3167-3178, 2001.

Tuck, A.F., Production of nitrogen oxides by lightning, *Q. J. R. Meteorol Soc.*, *102*, 749-755, 1976.

Uman, M.A., *The lightning discharge*, Academic, San Diego, Calif., 1987.

Wang, Y., A. W. DeSilva, C. G. Goldenbaum, and R. R. Dickerson, Nitric oxide production by simulated lightning: Dependence on current, energy, and pressure, *J. Geophys. Res., 103,* 19,149-19,159, 1998.

Williams, E.R., R. Zhang, and J. Rydock, Mixed phase microphysics and cloud electrification, *J. Atmos. Sci., 48,* 2195- 2203, 1991.

Zel'dovich, Y.B., and Y.P. Raizer, *Physics of Shock Waves and High Temperature Hydrodynamic Phenomenon*, Academic, San Diego, Calif., 1966.

Zhang, R., N.T. Sanger, R.E. Orville, X. Tie, W. Randel, and E.R. Williams, Enhanced NO_x by lightning in the upper troposphere and lower stratosphere inferred from the UARS global measurements, *Geophys. Res. Lett., 27,* 685-688, 2000.

D.J. Boccippio, Global Hydrology and Climate Center, NASA-Marshall Space Flight Center, Huntsville, Alabama, 35805, USA.

D.W. Bond, R.E. Orville, and R. Zhang (corresponding author), Department of Atmospheric Sciences, Texas A&M university, College Station, Texas, 77843-3150, USA. (zhang@ariel.tamu.edu)

Guy Brasseur and Xuexi Tie, National Center for Atmospheric Research, Boulder, Colorado, 80307, USA.

Gary Huffines, Department of Engineering Physics, Air Force Institute of Technology, 45433, USA.

(Received November, 27, 2000; revised June 26, 2001; accepted July 6, 2001.)