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A global three-dimensional time-dependent lightning source of tropospheric NO_x

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Abstract. The spatial and temporal distribution for a global three-dimensional, time-dependent lightning source of NO_x is constructed from a general circulation model's (GCM) deep moist convection statistics [*Manabe et al.*, 1974; *Manabe and Holloway*, 1975], observations of cloud-to-cloud and intracloud lightning fractions and the vertical distribution of lightning discharge [*Proctor*, 1991], and empirical/theoretical estimates of relative lightning frequency resulting from deep moist convection over ocean and over land [*Price and Rind*, 1992]. We then bracket the annual global emission of NO_x from lightning between 2 and 6 Tg N/yr, with a most probable range of 3 to 5 Tg N/yr, by comparing tropospheric NO_x simulations from the Geophysical Fluid Dynamics Laboratory Global Chemical Transport Model with measurements of NO_x and/or NO_y in the mid and upper troposphere where lightning is a major, if not the dominant, source. With this approach, the global magnitude of the lightning source is constrained by observed levels of NO_x, while the temporal and spatial distributions of the source are under the control of the parent GCM. Although our lightning source is smaller than many previous estimates, it is still the major source of NO_x and NO_y in the mid and upper troposphere for a latitude belt running from 30°N to 30°S, an important contributor to summertime free tropospheric levels over the midlatitudes, and a major contributor, even in the lower troposphere, to the low NO_x and NO_y levels over the remote oceans.

1. Introduction

NO_x ($NO + NO_2$) plays a key role in tropospheric chemistry [*Levy*, 1971; *Chameides and Walker*, 1973; *Crutzen*, 1974]. With a short tropospheric lifetime (1-10 days) and a number of diverse and dispersed sources, NO_x mixing ratios range from tens of parts per billion by volume (ppbv) in regions of surface pollution to a few parts per trillion by volume (pptv) in the marine boundary layer (MBL) of the remote central Pacific and are only sparsely measured throughout most of the troposphere [e.g., *Logan*, 1983; *Fehsenfeld et al.*, 1988; M.A. Carroll and L. Emmons, private communication, 1995]. In the foreseeable future, we do not expect that available measurements and proposed measurement campaigns will allow us to generate the detailed global NO_x fields needed to study a wide range of questions in global tropospheric chemistry. Rather, we will have to rely on simulated global NO_x fields that have been generated by global chemical transport models (GCTM) and evaluated with existing NO_x observations. The key to such simulations, assuming a GCTM with adequate representations of transport, chemistry, and removal, is an accurate estimate of the magnitude, spatial distribution and temporal variability of tropospheric NO_x sources.

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The global distribution, timing, and magnitude of five of the six currently known NO_x sources are relatively well characterized. They are based on detailed emission inventories and, where possible, simulated nitrate deposition and reactive oxidized nitrogen species concentrations have been compared with observations from regions of the troposphere dominated by an individual source. A sample of the range of their estimated global magnitudes, along with the appropriate references, are given in Table 1. However, the global distribution and timing of the sixth source, NO_x emissions from lightning discharge, have only been crudely characterized [*Hameed et al.*, 1981; *Penner et al.*, 1991], and previous estimates of the global magnitude, as shown in Table 1, vary widely. These estimates, most of which trace back to a 1925 estimate of global flash frequency [*Brooks*, 1925] and either first-principles calculations of NO_x per lightning stroke or atmospheric and/or laboratory measurements of NO_x in a lightning discharge, range from a few teragrams nitrogen per year up to unrealistic values exceeding 100 Tg N/yr (see Table 2 of *Lawrence et al.*, [1995] for more detail).

After an analysis of previous global source estimates, based both on direct calculations and in situ observations, *Liaw et al.* [1990] proposed 81Tg N/yr as the best estimate of the global lightning source. A recent study by *Lawrence et al.* [1995] reexamined the direct calculations and arrived at a much lower range of 1-8 Tg N/yr, though it should be noted they were extrapolating globally from a relatively limited number of local measurements.

Table 1. Global Sources of NO_x

Source	Estimated Magnitude, Tg N/yr	Reference
Fossil fuel	22	*Hameed and Dignon* [1988]
	21.2	*Levy and Moxim* [1989]
	22.2	*Benkovitz et al.* [1996]
	21.4	this work
Subsonic aircraft	1	*Beck et al.* [1992]
	0.45	*Wuebbles et al.* [1993]
Stratosphere	0.64	*Kasibhatla et al.* [1991]
Biomass burning	~6	*Hao et al.* [1989]
	8.5	*Levy et al.* [1991]
	5.8	*Penner et al.* [1991]
Soil biogenic emissions	~5	*Dignon et al.* [1992]
	4.7	*Müller* [1992]
	5.5	*Yienger and Levy* [1995]
Lightning	2.1	*Hameed et al.* [1981]
	19.1 - 152	*Liaw et al.* [1990]
	3 - 10	*Penner et al.* [1991]
	220	*Franzblau and Popp* [1989]
	1 - 8	*Lawrence et al.* [1995]
	2 - 6	this work

In an earlier global nitrogen budget study, *Logan* [1983] estimated that measurements of nitrate deposition in remote locations far from all other NO_x sources would support a lightning contribution of no more than 20 Tg N/yr. In their GCTM simulations for January and July, *Penner et al.* [1991] arrived at a similar upper limit. They used a 10°x15° lightning distribution based on observed thunder days and a formula relating, as a function of latitude, thunder days to lightning strokes, though it should be noted that lightning strokes/thunder day can be highly variable. They found that a lightning source of 3-10 Tg N/yr gave reasonable agreement with some observed free tropospheric levels of NO, though they appear to underestimate the upper tropospheric NO maxima observed by *Davis et al.* [1987] off the coast of California and by *Drummond et al.* [1988] over the subtropical and tropical Atlantic. A preliminary GCTM study by *Levy et al.* [1992], which used a very limited set of NO and NO_y observations in the upper troposphere, bracketed the global lightning source between 2 and 4 Tg N/yr.

Both previous GCTM studies and the *Lawrence et al.* [1995] reanalysis support a global lightning source that is at the low end of past estimates and is approximately 10% of current global NO_x emission estimates of ~40 Tg N/yr [e.g., *Moxim et al.*, 1994]. However, this emission of NO_x into the mid and upper troposphere, where it has a relatively long lifetime and other in situ sources are less than 1 Tg N/yr, has the potential to significantly influence the NO_x levels and the resulting ozone chemistry of that region [*Lin et al.*, 1988; *Trainer et al.*, 1991; A.A. Klonecki, private communication, 1995].

Not only have there been a very wide range of estimated global source strengths, but no ab initio calculations exist to generate a believable three-dimensional, time-dependent lightning source of NO_x. All previous approaches, as well as this study, combine numerical models, theory, and observations in the construction of an empirical source. One general approach [e.g., *Price and Rind*, 1992] uses a general circulation model (GCM) to construct global time-dependent maps of lightning and then uses theory and laboratory studies of NO_x emission per lightning flash to generate the global source of NO_x. We have taken a more empirical path. In section 2 our GCM is used to construct global time-dependent maps of deep moist convection, and observations and theories of intracloud (IC) and cloud-ground (CG) lightning are then used to convert the deep moist convection fields into three-dimensional, time-dependent maps of lightning. In section 3, rather than estimating the NO_x production per lightning flash from theory and observation, we determine the range of global scaling factors that produces the best fit between our GCTM simulations of NO_x and NO_y and observations from regions where lightning is expected to be a major, if not the dominant, contributor. This range of empirically determined global scaling factors then provides our estimated range of global source strengths for the emission of NO_x by lightning. The reactive nitrogen GCTM, employing a single scaling factor based on a global source strength of 3 Tg N/yr, is then used in section 4 to quantify the contribution of lightning to the levels of NO_x and NO_y throughout the troposphere.

2. Relative Global Source

Our construction of the relative global lightning source of NO_x has three stages. In section 2.1 a horizontal time-dependent distribution of deep moist convection is constructed using GCM statistics [*Manabe and Holloway*, 1975]. In section 2.2 we use observed vertical distributions of lightning origination and relative intracloud and cloud-ground frequency [*Proctor*, 1991] and an empirical ratio of lightning flashes in continental and maritime convection [*Price and Rind*, 1992] to convert the two-dimensional distribution of deep moist convection to a three-dimensional distribution of IC and CG lightning. The conversion of our lightning distribution to a relative distribution of NO_x emission then requires a number of simplifying approximations and estimates of the energy in CG and IC flashes.

2.1. Horizontal and Temporal Distribution of Lightning

Our horizontal and temporal distributions of lightning begin with simulated moist convection statistics, which are sampled once every 6 hours from a year's integration of a GCM with a horizontal resolution of ~265 km, 11 vertical levels (standard pressures of 990, 940, 835, 685, 500, 315, 190, 110, 65, 38, and 10 mbar), and no diurnal cycle [*Manabe et al.*, 1974; *Manabe and Holloway*, 1975]. While the model's resolution cannot resolve individual convective cells or even large mesoscale weather systems and their associated high-frequency lightning [e.g., *Goodman and MacGorman*, 1986], it will be shown that the model's seasonally varying regional distribution of convection adequately captures the observed global patterns of lightning. For the model's convective event to generate lightning, we require that saturated convection occur contiguously from ~800 mbar to at least 400 mbar. In this way we guarantee that the resulting NO_x emission by lightning will be directly coupled to the strong convective transport and mixing in the model.

Guided by the empirical analysis of *Price and Rind* [1992], deep moist convective events over the ocean, which are much less likely to generate lightning, are weighted by 0.1, except for the grid boxes containing the Caribbean Islands and Indonesia, which are treated as land points and given a weight of 1.0. Because of the model's lack of summertime precipitation over the southeastern U.S. and parts of eastern Asia and Australia, these regions, which are convective and which are known to generate a great deal of summertime lightning in the real world, do not satisfy our criteria (see the preceding paragraph) for convectively generated lightning in the model. Therefore we supplement moist convective events with dry convection in those regions. These supplemental dry convective events, which amount to ~4% of the total convective events sampled during the year, are then scaled by 0.75 to improve the agreement between the model's distribution of convection and the observed latitudinal distributions of lightning.

Implicit in relating deep moist convection frequency directly to lightning frequency is the assumption that all deep convective events, after the various empirical scalings, have the same flash intensity. Clearly, this is not strictly correct, as a number of theoretical and empirical studies have shown [see *Price and Rind*, 1992, and references therein], but it is a necessary simplification for this study. In the future, a logical extension would be to relate the flash intensity to the intensity of the individual convective events.

In Figure 1 the yearly averaged latitudinal distribution of the GCM's saturated deep convection frequency is compared with the observed yearly averaged latitudinal distribution of lightning flashes assembled by *Turman and Edgar* [1982] for dawn and dusk and by *Orville and Spencer* [1979] for dusk and midnight. In order to focus on the relative latitude distributions and to allow a direct comparison between saturated deep convection frequency and flash frequency, all three distributions are scaled to give the same number for the 0°-10°N band. Given the prevalence of year-round convection in the tropics and strong summer convection over the continents in the summer, the observed distributions in Figure 1 come as no surprise. Both measured and simulated distributions are concentrated between 20°N and 20°S with significant secondary contributions between 20°-40°N and 20°-40°S, a sharp drop-off poleward of 40°, and almost nothing poleward of 60°. In general, the simulated convective frequency falls between the two observed lightning distributions and captures the observed latitudinal distribution.

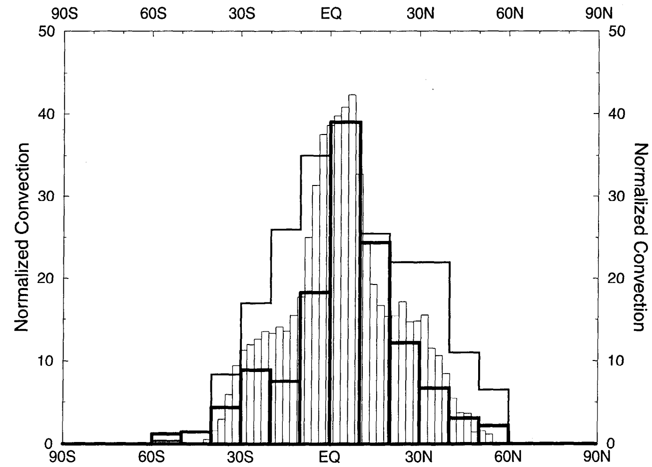

Figure 1. The simulated annual latitudinal distribution of deep convection is represented by the narrow bars. The heavy wide bars are the *Orville and Spencer* [1979] observations of annual latitudinal distribution of lightning flashes, and the thin wide bars are the *Turman and Edgar* [1982] observations. All three distributions have been scaled to give the same number for the 0°-10°N band.

In Figure 2 we show the winter, summer, and annual simulated latitude-longitude distributions of saturated deep convection frequency, which are considerably more complex than those in Figure 1. Note that the marine deep convection has been scaled by 0.1. The convection/lightning, which is clearly not uniformly distributed for a given latitude, is concentrated over land with a smaller, though not insignificant contribution over the tropical ocean. At midlatitudes the continental-based lightning occurs almost exclusively in the summer. In the tropics, the lightning migrates with the tropical rainy season over South America and Africa and with the movement of the Asian monsoon from the Indian subcontinent and Southeast Asia in the summer to Indonesia and northern Australia in the winter. Over the ocean, there are weaker maxima stretching down to 30°S in the western South Pacific throughout the year, off the east coast of the United States and Asia in the summer, and over the tropical Indian Ocean. A weak band of lightning observed over the wintertime North Pacific and Atlantic (S.J. Goodman, private communication, 1996) is not present in the simulation. While there are significant year-to-year variations in regional lightning intensity (see *Goodman and Christian* [1993] for a recent discussion), these simulated spatial patterns and their relative intensities are generally in good agreement with the seasonal and annual global lightning frequency maps of *Orville and Henderson* [1986], *Turman and Edgar* [1982], and *Goodman and Christian* [1993].

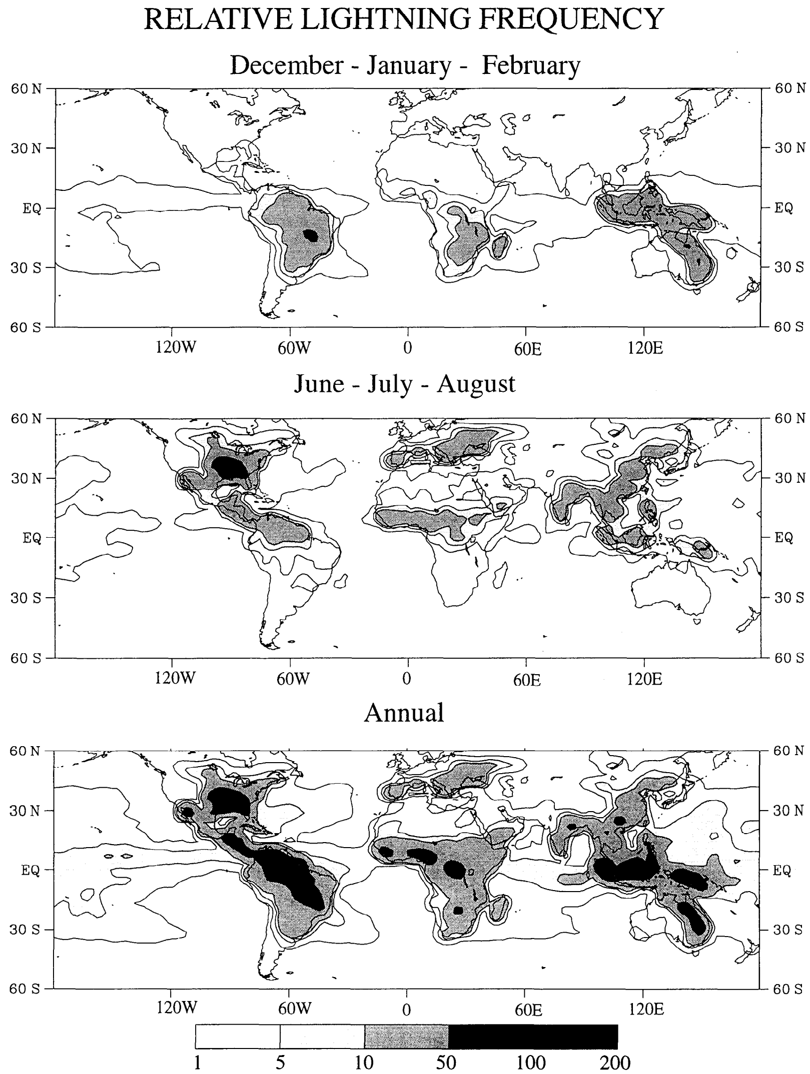

Figure 2. The simulated summer, winter, and annual distribution of deep convection, based on a once every 6 hours sample of the model's convection statistics. The actual numbers represent the number of 6-hour periods with convection occurring. This number can be linearly related to flash frequency if all deep convective events are assumed to have the same number of flashes.

2.2. Vertical Distribution of Lightning and NO_x emission

To determine the vertical distribution of NO_x emission for a given event, we must first generate the vertical distribution of intracloud and cloud-to-ground lightning. While it may be possible to do so from physical properties predicted by those GCMs with subgrid-scale parameterizations of clouds and moist convection [e.g., *Price and Rind*, 1993], we have chosen an empirical route. Our vertical distribution is based on the analysis of *Proctor* [1991], who monitored 773 lightning flashes in South Africa and determined both the vertical distribution of lightning flash origin and the vertical distribution the CG and IC fractions. The 13 thunderstorms over a 10-year period yielded a clear bimodal distribution with flash origination maxima at ~500 mbar and ~300mbar and a CG fraction of 0.28. The CG flashes originated almost completely in the lower maximum (~500 mbar), and the IC flashes, while distributed throughout, occurred primarily in the upper maximum (~300mbar). Contrary to a number of previous studies that argued for a strong latitude dependence in the CG fraction, a recent analysis of cloud and lightning data by *Price and Rind* [1993] argues that the fraction varies only between 0.24 and 0.28 over the latitude range of 0°- 40°. Therefore we have chosen to use the *Proctor* [1991] distributions of IC and CG for all latitudes.

For the final conversion to a time-dependent, three-dimensional source of NO_x, we make two further simplifications: All CG lightning flashes produce the same amount of NO_x (CG_{NOx}), which is linearly apportioned among the levels according to their mass; NO_x production by IC lightning (IC_{NOx}) is also the same for all flashes and remains within the model layer of origination. The approximation of the same NO_x production for all IC and for all CG flashes is related to the earlier approximation of the same flash frequency for all convective events and results from the same lack of model-generated convective intensity data. There are also two implied assumptions: NO_x production varies linearly with the flash energy, and NO_x production is independent of pressure over the range 1000 mbar-200 mbar. *Holmes et al.* [1971] found that CG flashes contained at least 3 times the energy of

IC flashes, and *Kowalczyk and Bauer* [1982] adopted a factor of 10, which gives an NO_x flash yield in agreement with both theory and laboratory measurements [*Lawrence*, 1995]. Recently, on the basis of calculations by *Cooray* [1996] that find similar levels of energy dissipation in CG and IC flashes, L. Gallardo (private communication, 1995) has argued that CG and IC flashes produce similar levels of NO_x.

Two vertical distributions for the percentage production of NO_x are shown in Figure 3. Case a, which was used in the preliminary study by *Levy et al.* [1992], has $IC_{NOx} = CG_{NOx}$. IC flashes dominate NO_x production and maximize NO_x emission in the upper troposphere with 48% above 7 km, 42% between 7 and 2 km, and only 10% below 2 km. Case b, which has $CG_{NOx} = 10xIC_{NOx}$, will be used in our current calculations. It produces a maximum NO_x emission in the middle troposphere with only 16% above 7 km, 54% between 7 and 2 km, and 30% below 2 km. While case b will deposit ~3 times more NO_x in the continental boundary layer, where it may be subject to rapid loss, both distributions emit most of the NO_x in the free troposphere. This fact, when coupled with lightning's occurrence in a region of upward vertical motion, greatly reduces the impact of the factor of 10 change in the relative NO_x production by IC and CG lightning.

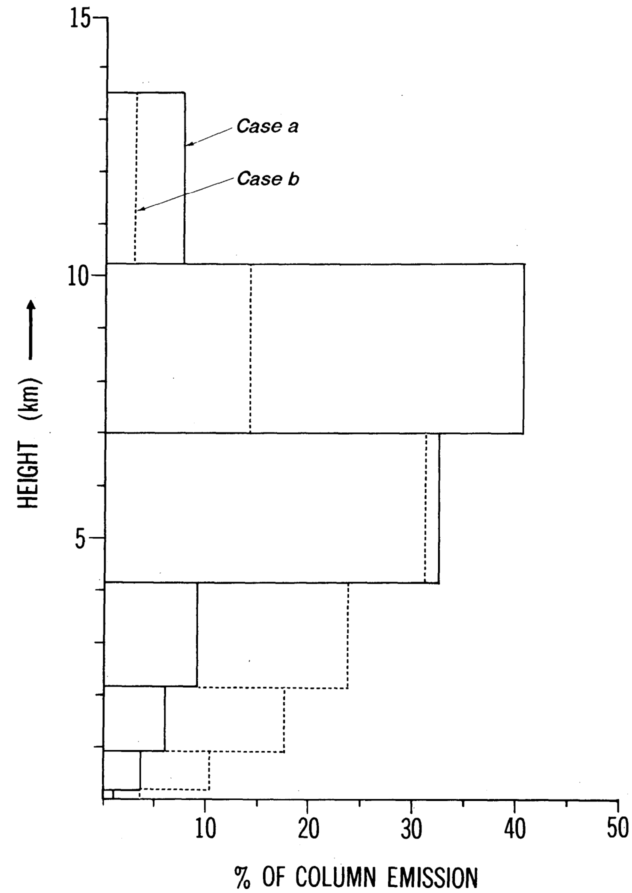

Figure 3. Vertical distribution of the lightning production of NO_x during deep convection. The dashed line represents case b with CG NO_x production = 10xIC NO_x production. The solid line is for case a with CG = IC.

3. Absolute Global Source

Having generated our three-dimensional, time-dependent relative distribution of NO_x emissions by lightning, we still need to quantify the yearly global source strength. We do so by comparing NO_x and NO_y simulated by our GCTM with available observations from regions, primarily the middle and upper troposphere between 30°S and 30°N, that are dominated or at least strongly affected by emissions from lightning. Before examining the data comparison and the calculations of the global lightning source in section 3.2, we should briefly discuss our reactive nitrogen GCTM.

3.1. GCTM Description

The Geophysical Fluid Dynamics Laboratory (GFDL) GCTM is driven by 12 months of 6-hour time-averaged wind, temperature, and precipitation fields from a GFDL GCM [*Mahlman and Moxim*, 1978]. The GCTM has the same horizontal and vertical resolution as the parent GCM and includes parameterizations of horizontal subgrid-scale transport and vertical mixing by dry and moist convection (for details, see Appendix A of *Levy et al.* [1982], and section 2 of *Kasibhatla et al.* [1993]).

The GCTM explicitly treats the transport and chemistry of three tracers: nitrogen oxides ($NO + NO_2 + NO_3 + N_2O_5$), nitric acid ($HNO_3$), and peroxyacetylnitrate (PAN). Chemical interconversion rates among the three tracers, as well as intraconversions among the nitrogen oxides, are calculated off-line and are carried in the GCTM as temporally varying, two- and three-dimensional fields (see section 2 of *Kasibhatla et al.* [1993] and section 2 of *Moxim et al.* [1996] for details). The one exception is the thermal decomposition rate of PAN, which is calculated on-line in the model as a function of the local temperature. Dry deposition fluxes of NO_x, HNO_3, and PAN over land and of HNO_3 over oceans, ice, and snow assume a balance between surface deposition and the turbulent flux in the bottom half of the lowest model level [see *Kasibhatla et al.*, 1993, equation 2.2] and use measured deposition velocities of individual reactive nitrogen species (see section 3 of *Kasibhatla et al.* [1993] for details). The removal of HNO_3 by precipitation is calculated using the local precipitation rate, and the wet removal tendency is proportional to the local tracer mixing ratio (see section 2 of *Kasibhatla et al.* [1991] for details).

Along with the natural lightning source of NO_x being developed in this paper, the model incorporates five other sources: (1) emissions of 21.4 Tg N/yr from surface fossil fuel combustion [*Levy and Moxim* [1989] updated with the GEIA Project [*Benkovitz et al.*, 1996]; (2) aircraft traffic emissions of 0.45 Tg N/yr [*Kasibhatla*, 1993; *Wuebbles et al.*, 1993]; (3) biomass-burning emissions of 8.5 Tg N/yr [*Levy et al.*, 1991], updated by a reduction factor of 0.7 for emissions from the savannas of northern Africa [*Delmas et al.*, 1995]; (4) primarily natural NO_x emissions from soil of 5.5 Tg N/yr [*Yienger and Levy*, 1995], updated by a reduction factor of 0.1 for emissions from the savannas of northern Africa [*Le Roux et al.*, 1995]; and (5) the injection of 0.65 Tg N/yr of primarily natural stratospheric NO_y from the stratosphere [*Kasibhatla et al.*, 1991].

3.2. Empirical Global Scaling Factor

In order to determine the global empirical scaling factor, we need global simulations of reactive nitrogen species for two source conditions: (1) a simulation employing all five nonlightning sources of tropospheric NO_x; and (2) a second simulation that produces NO_x and NO_y levels from the lightning source, scaled to an arbitrary global amount, in our case 1 Tg N/yr.

The determination of the global scaling factor is summarized in Table 2. From left to right are the observed values for NO_x or NO_y from regions where lightning plays an important role, the simulated values for the same locations using the five nonlightning sources of NO_x, the residual (observed minus simulated five source) values that must be explained by lightning, the simulated contribution from a 1 Tg N/yr global lightning source, and the actual global lightning source required to bring a complete six-source simulation into agreement with the observations. We generate median values for the detailed observations in Table 2, while for the sparser data we simply give the range of the data (depicted by brackets). The binning of observed data by altitude was dictated by the model's vertical standard pressure layers as follows: 190 mbar implies a range of 150 to 241 mbar, 315 implies 241-412 mbar, and 500 implies 412-607 mbar. The simulated median values were generated by sampling all the grid boxes that a flight path passed through over the given time period of the expedition.

The first three missions, STEP, STRATOZ III and CITE 2 (acronyms are identified in the footnote to Table 2), were the only data sets available when *Levy et al.* [1992] used the case a vertical distribution to arrive at a preliminary estimate of 1-4 Tg N/yr for the global lightning source. With the case b vertical distribution, which is a lower limit on the emission of NO_x in the upper troposphere, the preliminary estimated range would have risen to 2-6 Tg N/yr. Recently, extensive aircraft measurements of NO_x from the TRACE-A, PEMwest-A and New Mexico missions have become available, and we find, with the exception of the 315 mbar data over New Mexico, that they all require a global lightning source in the range of 3-6 Tg N/yr. The large majority of the observations require a source in the range of 3-5 Tg N/yr. A more detailed examination of the 315 mbar data over New Mexico shows a bimodal distribution with a small peak at 60-100 pptv and a larger peak at 180-240 pptv. Clearly, the

Table 2. Estimated Global Lightning Emissions of NO_x and NO_y

Mission[a] and Location	Month	Pressure, mbar	Species	Observed, pptv	Simulated,[b] pptv	Observed Minus Simulated,[a] pptv	Lightning Contribution, pptv/(TgN/yr)	Required Lightning Source, TgN/yr
STEP,[c] Darwin, Australia	Jan.	200	NO_y	[340 - 420]	119	[221-301]	77	[2.8 - 3.8]
STRATOZ III[d] Dakar, Senegal	June	190	NO_x	[100 - 175]	61	[39 - 114]	27	[1.4 - 4.2]
CITE2,[e,f] east N. Pacific (~40°N)	Aug.	500	NO_x	31	13	18	3	6
			NO_y	298	219	79	36	2.2
TRACE-A,[g] S. Atlantic (0°-30°S)	Oct.	190	NO_x	200	69	131	25	5.3
		315	NO_x	134	56	78	18	4.3
		500	NO_x	66	38	28	7	4
PEMwest-A[h] west N. Pacific (0°25°N)	Oct.	190	NO_x	62	20	42	12	3.5
		315	NO_x	51	9	42	8	5.25
		500	NO_x	26	11	15	4	3.75
New Mexico[i]	July, Aug.	190	NO_x	293	237	56	15	3.8
		315	NO_x	215	48	167	8	21
		500	NO_x	84	26	58	10	5.8

[a] Mission acronyms are STEP, Stratosphere-Troposphere Exchange Project; STRATOZ, Stratospheric Ozone Experiment; CITE, Chemical Instrumentation Test and Evaluation; TRACE-A, Transport and Atmospheric Chemistry Near the Equator-Atlantic; PEMwest, Pacific Exploratory Mission-west.
[b] Simulated values for the five nonlightning sources of NO_x.
[c] Murphy et al. [1993].
[d] Drummond et al. [1988].
[e] Carroll et al. [1990].
[f] Hübler et al. [1992].
[g] Smyth et al. [1996].
[h] Bradshaw (private communication, 1994).
[i] Ridley et al. [1994].

simulation misses the higher peak. This may be due to a model deficiency in lightning in the region, a particular transport situation that is not properly simulated, or aliased data due to limited sampling.

3.3. Perspective on Uncertainty

While we have chosen a most probable range of 3-5 Tg N/yr, there are a number of uncertainties in both the analysis and the actual measurements that could both raise and lower the estimated global source strength.

First, there is the suspicion [*Crawford et al.*, 1996; B. Ridley, private communication,1995; J. Bradshaw, private communication, 1995; D.D. Davis, private communication, 1995] that a number of the NO_x measurements in the upper troposphere, because of discrepancies between observed and theoretical NO/NO_2 ratios, may be an overestimate. As a check, we have compared our simulations with calculated NO_x from the PEMwest-A expedition that are based on measured NO and O_3 [*Crawford et al.*, 1996]. We find that while these NO_x values give lower estimates of the global source strength (~3 Tg N/yr), they still fall in the same general range.

A potentially significant source of uncertainty is our use of NO_x, with its implicit reliance on our calculated OH fields, to empirically scale the source. For example, a systematic 50% increase in the upper tropospheric OH mixing ratios would lead to an approximate factor of 2 increase in the required global lightning NO_x source (6-10 Tg N/yr). While we have previously compared our OH-based CH_3CCl_3 atmospheric lifetime to the observed value (see *Kasibhatla et al.* [1993] for

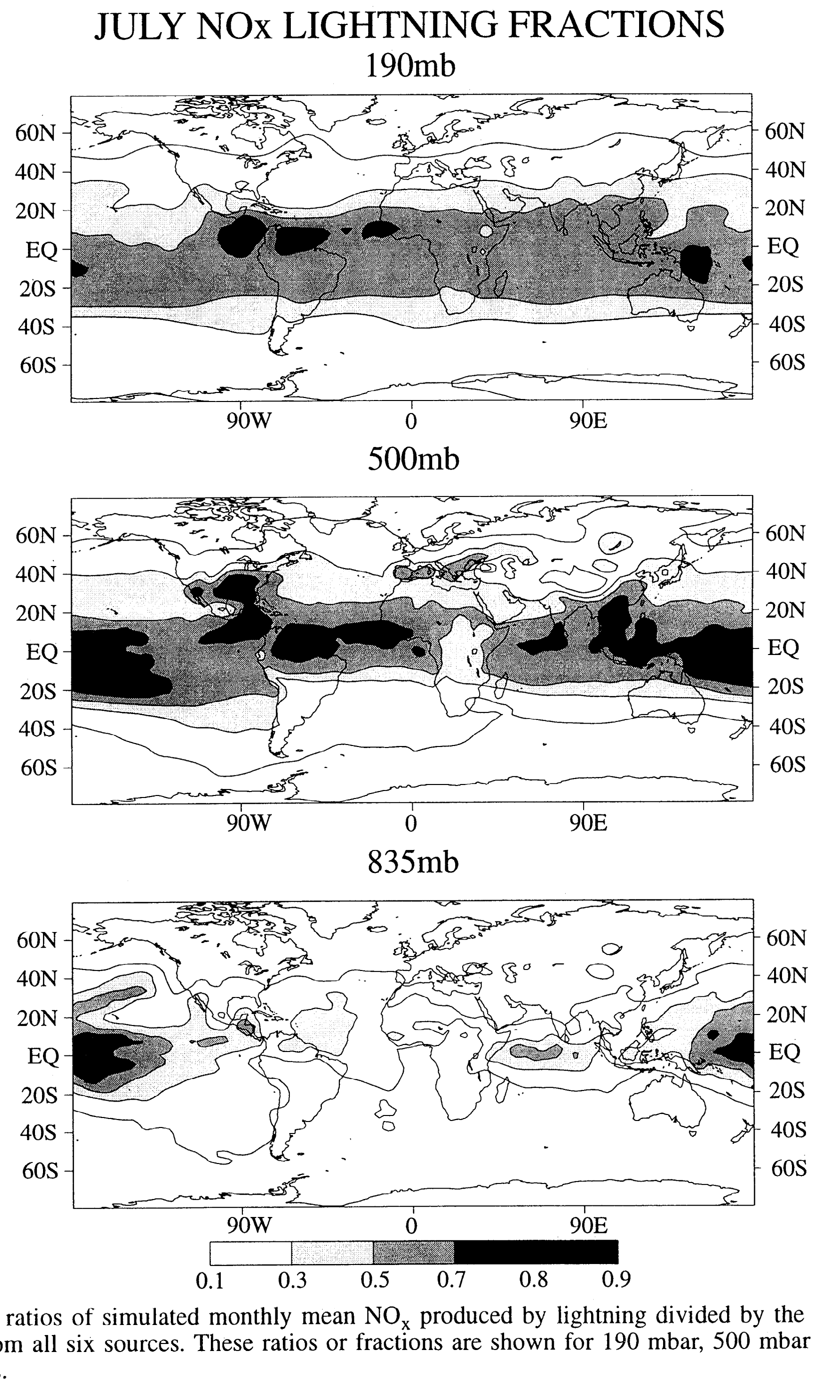

Figure 4. July ratios of simulated monthly mean NO_x produced by lightning divided by the monthly mean NO_x levels from all six sources. These ratios or fractions are shown for 190 mbar, 500 mbar and 835 mbar pressure levels.

details), this applies primarily to the lower half of the troposphere because of the large activation energy for CH_3CCl_3 oxidation. However, our upper tropospheric OH is in reasonable agreement with other calculations [*Kasibhatla et al.*, 1991].

While the use of NO_y observations, rather than NO_x, for the empirical scaling would avoid most of the OH uncertainty, there is currently a great deal of uncertainty in the aircraft measurements of NO_y [*Crosley*, 1996]. If the sum of the principal measured species (NO_x + HNO_3 + PAN) is used, the resulting range (1.5-6.5 Tg N/yr) is quite similar to that determined with NO_x. On the other hand, using directly measured NO_y leads to a much wider and less certain range of values with a range of 0-18 Tg N/yr and a mean and standard deviation of 10 Tg N/yr and 6 Tg N/yr, respectively.

Another source of uncertainty is the model's simulation of convective transport (for details, see Appendix A of *Levy et al.* [1982], and section 2 of *Kasibhatla et al.* [1993]). If the model underestimates convective transport from the atmospheric boundary layer (ABL), it will also underestimate the contribution of the large surface sources to NO_x levels in the upper troposphere. The correct convective transport would then require a reduction in the already small (3-5 Tg N/yr) global lightning source. If, on the other hand, convective transport is overestimated, the reverse is true. As an upper limit, we could assume that none of the surface sources contribute to the NO_x levels in Table 2. This could raise the range of the global source strength as high as 4-9 Tg N/yr.

Although lightning has a strong diurnal dependence in the real world, our simulated source has no diurnal structure. However, by tying our simulated NO_x emission to the model's vertical instability, which is observed to be highly correlated with lightning in the real world, we do keep the NO_x emission strongly correlated with the model's upward vertical motion

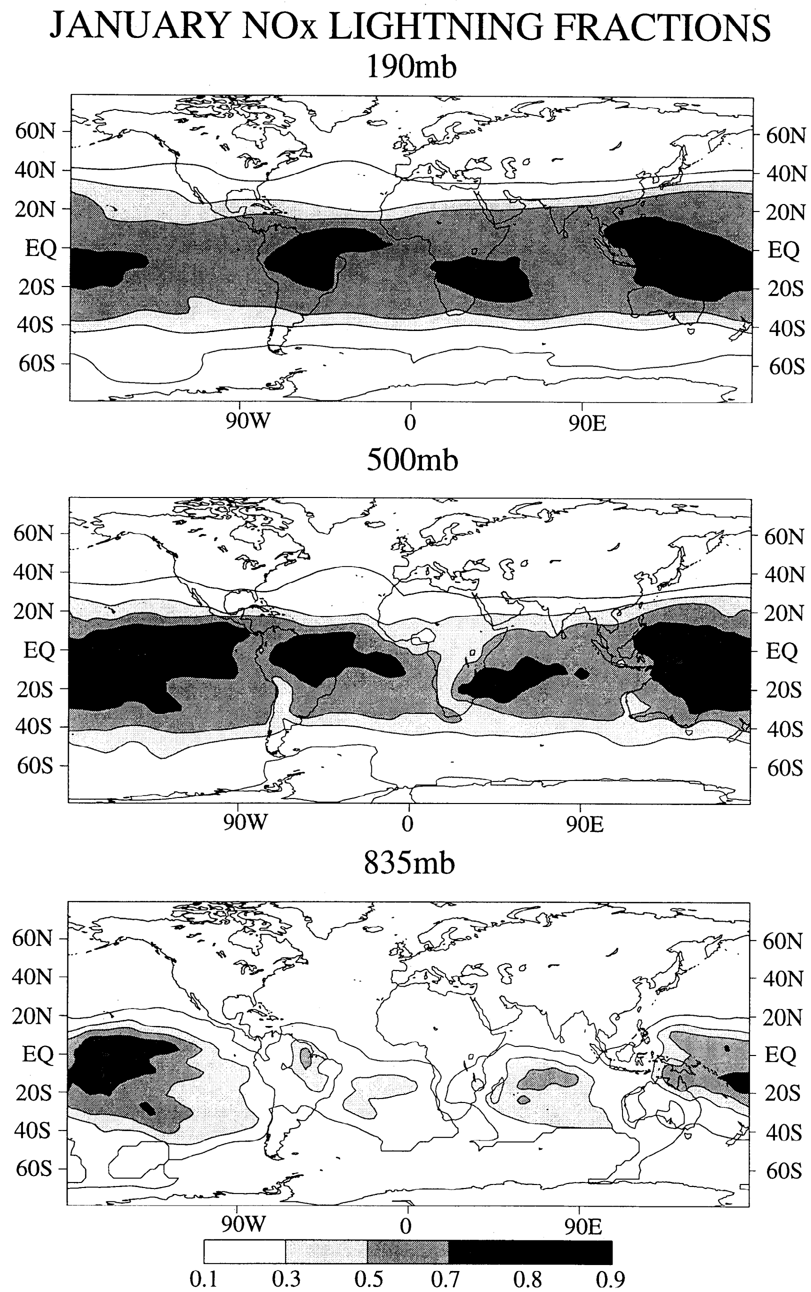

Figure 5. The same ratios as in Figure 4, except for January monthly means.

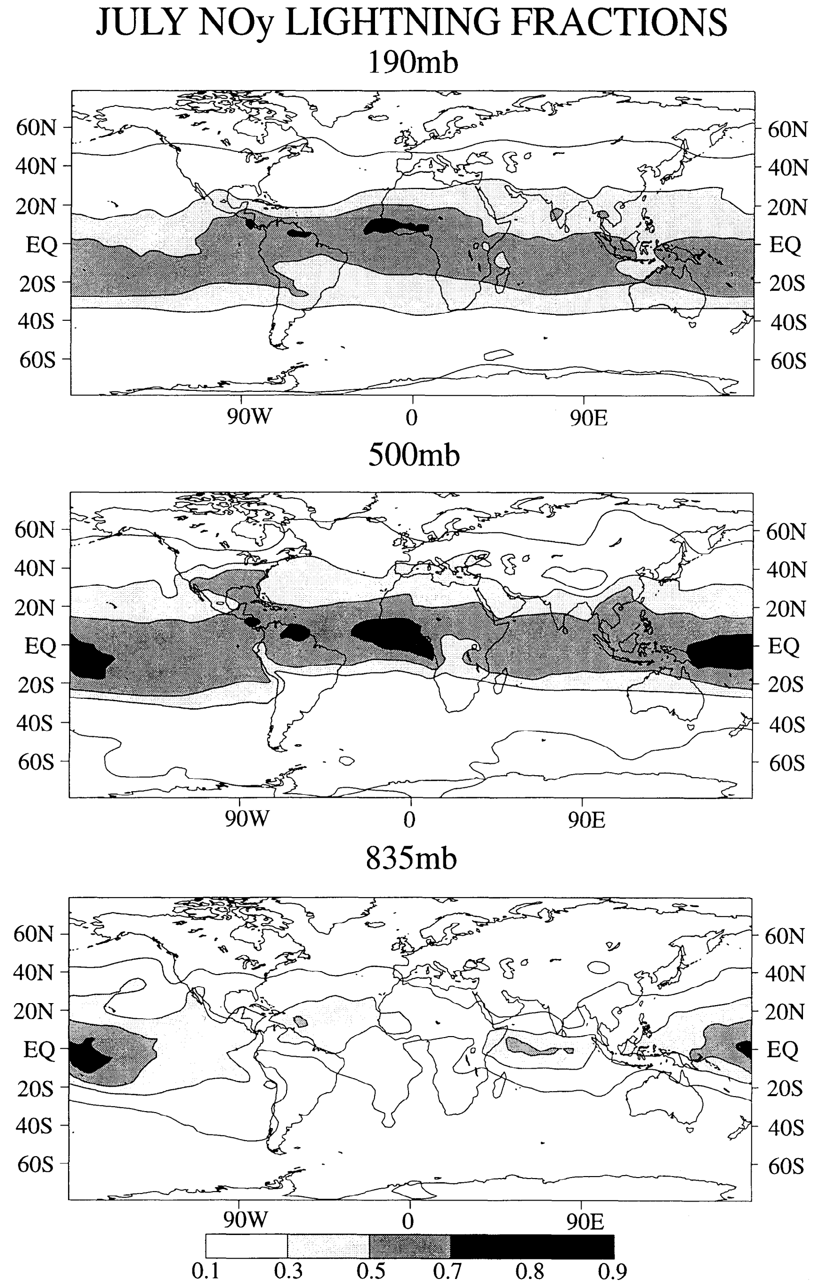

Figure 6. July ratios of simulated monthly mean NO_y produced by lightning divided by the monthly mean NO_y levels from all six sources. These ratios or fractions are shown for 190 mbar, 500 mbar and 835 mbar pressure levels.

and mixing. A previous simulation, which ignored this strong correlation between lightning and upward vertical motion, underestimated NO_x in the upper troposphere by 30% or more. While both the neglected diurnal fluctuations in temperature and systematic errors in the model's temperature field may affect PAN lifetimes in the upper troposphere, PAN chemistry itself has only a small impact on NO_x levels in that region [*Moxim et al.*, 1996].

Considering all these possible uncertainties, it would appear that our analysis arrives at a clear upper limit of 10 Tg N/yr for the global source strength. Taking a completely different approach, *Lawrence et al.* [1995] have arrived at a range of 1-8 Tg N/yr for the global source of lightning NO_x. They combine a reanalysis of in situ observations and laboratory studies of NO_x emissions by lightning flashes with a relatively crude estimate of global flash frequency. In a third approach, C. Price (private communication, 1995) uses International Satellite Cloud Climatology Project (ISCCP) observed clouds and theoretical and empirical relations between cloud properties and flash frequency to derive detailed maps of global flash frequency. These are then combined with a relatively crude estimates of NO_x production by lightning flashes to arrive at a global budget of 10-15 Tg N/yr. Just recently, *Kumar et al.* [1995] combined satellite observations of global thunderstorm activity and the formula for NO_x production per flash developed by *Borucki and Chameides* [1984] to arrive at a global source of 2 Tg N/yr. All the approaches have different weaknesses and different strengths. Nonetheless, all give global budgets in the same general range (1-15 Tg N/yr) with enough uncertainty to explain much of the remaining difference.

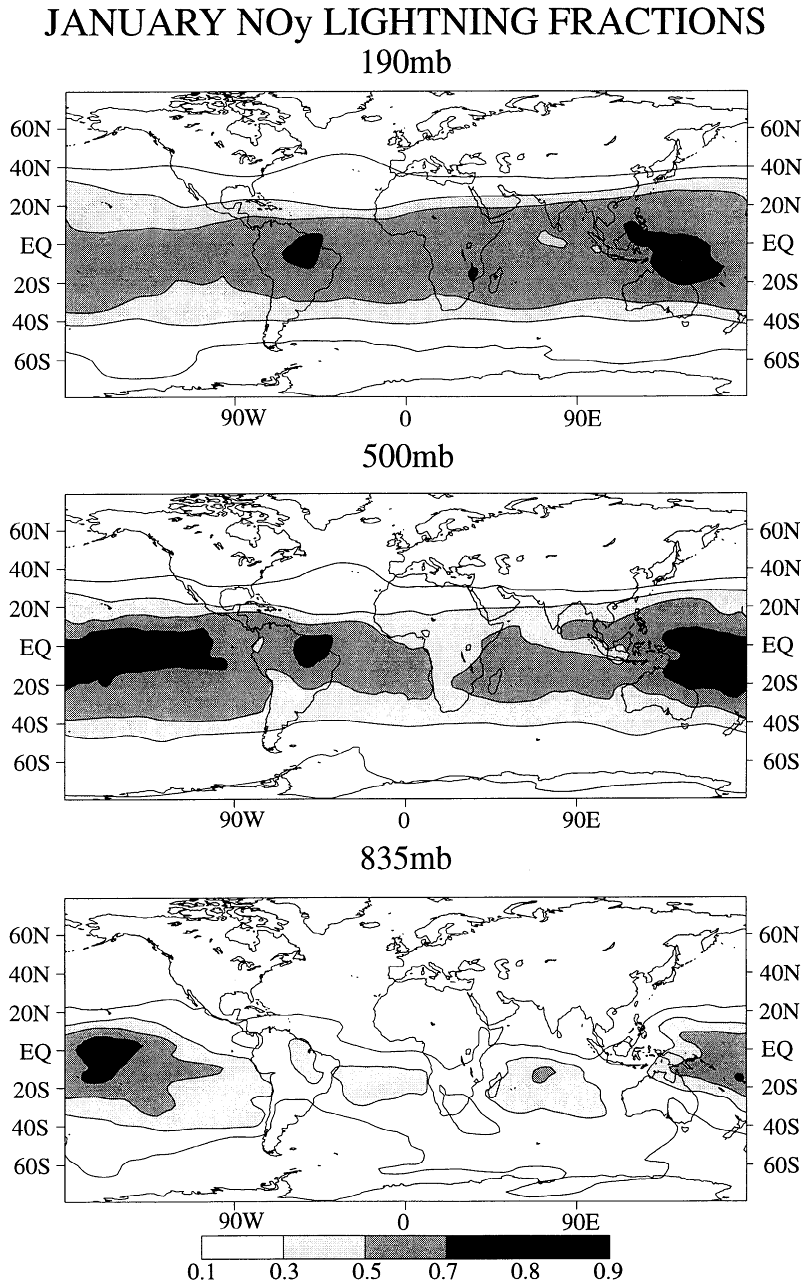

Figure 7. The same ratios as in Figure 6, except for January monthly means.

4. Impact of Lightning on Tropospheric NO_x and NO_y [$NO_x + HNO_3$ + PAN]

While lightning flashes may only provide ~10% of the ~40 Tg N/yr of NO_x currently emitted into the troposphere, they release NO_x into the free troposphere, where it has a relatively long chemical lifetime and can be rapidly transported. This magnifies its impact relative to the competing surface emissions. In Figures 4 and 5 we show global 190 mbar, 500 mbar and 835 mbar maps of the fraction of simulated NO_x produced by lightning for July and January, respectively. We chose a conservative lower limit of 3 Tg N/yr for the global lightning source strength in these calculations.

Throughout the tropics and subtropics, lightning is the major source of NO_x in the upper half of the troposphere, while still making a significant contribution to NO_x levels in the lower troposphere over the remote oceans, where NO_x levels are already very low (< 10 pptv). It is the most dominant in the middle troposphere, particularly during July. In the upper half of the troposphere, lightning makes significant contributions up to 40°N and 40°S in their respective summers, is a particularly important source over the continents during summertime convection, and contributes a major fraction over the remote oceans throughout the year. While signs of the convectively active regions appear in the upper troposphere, the impact of lightning is relatively zonal as a result of both the relatively long chemical lifetime of NO_x in that region and the rapid transport by the stronger middle and upper tropospheric horizontal winds.

Fractions of simulated NO_y resulting from the lightning emissions are shown in Figures 6 and 7. They are qualitatively similar to the NO_x fractions shown in Figures 4 and 5 with lightning being the dominant source of NO_y in the tropical mid and upper troposphere and in the tropical lower troposphere over the oceans. However, the percentage of NO_y resulting from the lightning source is generally 10-20% smaller than the NO_x percentage in the tropical troposphere, particularly over the ocean, where lightning is the only in situ source. It appears that NO_y, primarily as PAN and HNO_3, is more readily transported from the surface sources to the free troposphere than is NO_x, while there is already a direct local source of NO_x in that region. Therefore we believe that any analysis of NO_x source contributions that focuses solely on NO_y will underestimate lightning's contribution to NO_x and therefore indirectly underestimate its impact on tropospheric ozone chemistry. It should be further noted that our simulations find lightning generally supplying less than 10% of the NO_x and NO_y in the troposphere poleward of 50°N and 50°S during both summer and winter.

5. Summary and Conclusions

By combining a GCM's convection statistics, empirical relationships and observations of lightning origination and CG : IC flash ratios, and some simplifying assumptions about NO_x formation by lightning, we are able to generate a "best-estimate" three-dimensional, time-dependent source function for lightning NO_x. Further, we find that a global source strength ranging from 2 to 6 Tg N/yr with a most probable range of 3-5 Tg N/yr is sufficient to explain most levels of NO_x measured in the middle and upper troposphere of the tropics and subtropics. With this approach, the global magnitude of the lightning source is constrained by observed levels of NO_x, while the temporal and spatial distributions of the source are under the control of the parent GCM. Some uncertainties in measured NO_2 in the upper troposphere would support the lower end of the range, while other uncertainties in simulated OH and convective transport could support an upper limit of 10 Tg N/yr. However, even for a global source of 3 Tg N/yr, we find that NO_x emissions from lightning are the dominant source of tropospheric NO_x and NO_y in the upper half of the tropical and subtropical troposphere.

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