Chapter 26
Lightning and NO_X Production in Global Models

Kenneth Pickering, Heidi Huntrieser and Ulrich Schumann

Abstract In the upper troposphere lightning is the major contributor to the production of nitric oxide, which is a critical precursor gas for ozone production. It is therefore important that this source is simulated with a high accuracy in global chemical transport models and global chemistry/climate models. This chapter reviews development of the parameterization of lightning-produced nitric oxide in such models and the various components required such as flash rate distribution, NO production per flash and its vertical distribution. The results from simulations with different global models, the uncertainties and the impact on ozone are discussed.

Keywords Lightning · Nitrogen oxides · Aircraft observations · Chemical transport modelling · Cloud-resolved modelling · Tropospheric ozone

26.1 Introduction

Nitric oxide (NO) is produced in very hot lightning channels through the Zel'dovich mechanism (Zel'dovich and Raizer 1967) due to oxygen (O_2) and nitrogen (N_2) dissociation. As the channels cool to 3000–4000 K, NO is formed in the resulting plasma and is "frozen in" during the subsequent cooling to ambient temperature. Within seconds NO is converted to NO_2 by reaction with ambient ozone (O_3) and – during daytime – photolysed back to NO. An equilibrium is reached after about 100 s known as the photostationary state. The sum of NO and NO_2 is referred to as NO_X.

The magnitude of the NO_X production per lightning flash and the total global production are still a matter of debate. Schumann and Huntrieser (2007) have provided a comprehensive summary of the current knowledge of lightning-NO_X (LNO_X) production and suggest that the total global production, is 2–8 Tg/a with a most likely value of 5 Tg/a. (Here and below, the production rate is given in terms of

K. Pickering (✉)
NASA Goddard Space Flight Center, Laboratory for Atmospheres, Greenbelt, MD, USA
e-mail: Kenneth.E.Pickering@nasa.gov

H.D. Betz et al. (eds.), *Lightning: Principles, Instruments and Applications*,
DOI 10.1007/978-1-4020-9079-0_26,

equivalent nitrogen mass per year.) They state that this amount represents approximately 10% of the total NO production from all sources, but a far larger share of the amount emitted into the upper troposphere. NO is the most critical precursor gas for photochemical ozone production in the troposphere. Therefore, accurate knowledge of NO from the lightning source is essential in estimating ozone production rates.

Tropospheric ozone is the third most important greenhouse gas (IPCC 2007) with an estimated global annual average radiative forcing of 0.35 W/m^2 with 0.25–0.65 W/m^2 uncertainty. Radiative forcing associated with ozone is most sensitive to changes in ozone in the upper troposphere and lower stratosphere. LNO_X production occurs primarily in the middle and upper troposphere, and some of that produced lower in the atmosphere is convectively transported upward. It is known that the O_3 production efficiency per NO_X molecule typically increases with height, due to the longer lifetime of NO_X in the upper troposphere compared to the boundary layer. The lifetime increases as the chemical destruction rate decreases which is the case in the upper troposphere due to the colder and drier environment. Therefore, LNO_X has the greatest potential to influence ozone production in the middle and upper troposphere, and especially in tropical regions where the flash rate and insolation are highest (see Fig. 18 in Schumann and Huntrieser (2007) from Grewe (2007)). Aside from the aircraft source with 0.7 Tg/a (Schumann and Huntrieser 2007), lightning is the only other source of NO_X emitted directly into the upper troposphere. Therefore, it is important that we improve our knowledge of the magnitude and location of LNO_X production.

An accurate representation of the LNO_X source strength and temporal and spatial distribution is needed in global chemical transport models (CTM) and global chemistry/climate models (GCM or CCM). Offline chemical transport models are typically driven by meteorological fields produced by global data assimilation systems (e.g., National Center for Environmental Prediction (NCEP), European Centre for Medium Range Forecasts (ECMWF), and Global Modeling and Assimilation Office (GMAO)). Fields of winds, temperatures, convective fluxes, and cloud cover from these assimilation systems are used in the calculation of advection and chemical transformations. In such models there is no interaction between the resulting chemical distributions and the meteorology. Some global climate models run with online chemistry allowing radiative feedback due to changes in concentrations of atmospheric trace gases or aerosols. This radiative forcing can affect the atmospheric dynamics in the model leading to perturbed circulation patterns.

This chapter will review efforts in representing the LNO_X source in such offline and online models. Specifically, it will review the research that has led to the development of the various components of LNO_X parameterizations required for such global models. These components include a method to specify the geographic and temporal distribution of flashes (Section 26.2), an amount of NO production per flash (Section 26.3), and a method of specifying the effective vertical distribution of LNO_X emissions, including the effects of convective transport (Section 26.4). Our final section describes the information concerning LNO_X gained from such global models, the uncertainties and its impact on O_3 (Section 26.5).

26.2 Flash Rate Distributions

Lightning has been monitored from ground-based networks in a number of countries and detected routinely from space by satellites since 1988 and 1995, respectively. Both types of systems have generated a wealth of data concerning lightning flash rates. However, these data cannot be used directly in global or regional models for several reasons. Injecting NO from lightning at the times and locations of observed flashes will not necessarily match in space and time with the convective transport of ozone precursors in the model. The resulting ozone photochemistry will not be accurate. Ground-based network data also cannot be used because the systems with most geographic coverage record mainly CG flashes and only a few of the IC flashes. The satellite data, which does record both CG and IC flashes, cannot be used because of the very small amount of coverage on a daily basis. Gridded climatologies of the Optical Transient Detector (OTD) and Lightning Imaging Sensor (LIS) data have been constructed (http://thunder.nsstc.nasa.gov/data). However, these data suffer from the same problem (mismatch with model convection) as any observational data set if they are used directly in a model. Therefore, parameterizations for estimating flash rates in the model must be employed. These typically use various meteorological variables from the driving meteorological assimilation or from the climate model as predictors of flash rate. This approach makes the assumption that the meteorological fields used to drive the model adequately represent the deep convection which leads to the occurrence of lightning.

A number of global CTMs have used the cloud-top height from the driving meteorological model for estimating lightning flash rates. Theoretical and observational formulation of a relationship between flash rate and the fifth power of the cloud height stems from early work by Vonnegut (1963) and Williams et al. (1985). Formal power laws for predicting flash rates for total lightning from cloud-top height were derived by Price and Rind (1992). Separate equations for continental and marine lightning were presented as follows:

$$F_c = 3.44 \times 10^{-5}\, H^{4.9} \qquad \text{continental}$$

$$F_m = 6.40 \times 10^{-4}\, H^{1.73} \qquad \text{marine}$$

where F is the total flash rate in flashes per minute and H is the cloud-top height in kilometers. These power laws were originally meant to apply for maximum flash rates and cloud-top height. However, they have been applied in many CTMs on an instantaneous basis. Limitations of this approach have been discussed by e.g. Ushio et al. (2001), Allen and Pickering (2002), and Boccippio (2002). Ushio et al. (2001) analyzed Tropical Rainfall Measuring Mission (TRMM) precipitation radar and LIS flash data to show that instantaneous cloud-top height and flash rates can be related. However, resulting best fit power law relationships varied considerably with location and season. Through comparison of model estimated flash rates (predicted using cloud-top height from the GEOS-STRAT meteorological assimilation) with OTD/LIS data Allen and Pickering (2002) noted that the Price and Rind scheme underestimated flashes over the oceans by a factor of seven, overestimated the flash rates over tropical South America, and underestimated tropical African flashes. Boccippio (2002) observed that the Price and Rind marine parameterization is inconsistent with Vonnegut's original theoretical framework, and suggested a revised scaling relation.

Observational work by Petersen and Rutledge (1998) has suggested regime-dependent relationships between flash rate and convective precipitation. Meijer et al. (2001) and Allen and Pickering (2002) derived relationships for predicting CG flash rate (flashes per minute) using convective precipitation (CP in mm day^{-1}) from the meteorological models driving chemical transport. Allen and Pickering reported that separate continental and marine formulas were needed as shown below. These formulas are applicable for $2^\circ \times 2.5^\circ$ grid cells using the GEOS-STRAT data. The CG fraction of the total flash rate was derived from the relationship to cold-cloud thickness of Price and Rind (1993).

$$F_{CGocean} = 0.0523 - 0.048CP + 0.00545CP^2 + 0.0000368CP^3 - 0.000000371CP^4$$

$$F_{CGland} = 0.0375 - 0.0476CP + 0.00541CP^2 + 0.000321CP^3 - 0.00000293CP^4$$

Stronger updrafts within a thunderstorm lead to greater charge separation within the cloud, which may lead to greater flash rates. Price and Rind (1992) discuss relationships between updraft velocity and flash rate, which were applied by Pickering et al. (1998). Allen et al. (2000) were the first to utilize the upward cloud mass flux (M in kg m^{-2} min^{-1}) from a convective parameterization in a global model as a predictor of flash rate. Through tests over the eastern USA and western North Atlantic, they found that separate marine and continental formulas were not needed when this predictor variable is used. Allen and Pickering (2002) formalized the relationship between flash rate and upward cloud mass flux (a fourth-order polynomial) and applied it globally. The flash rates (flashes per minute for a $2^\circ \times 2.5^\circ$ grid cell) predicted by this formula must be adjusted for grid cell size and scaled upward to account for IC flashes. In general, flashes over the oceans were better predicted than with the cloud-top height approach, but flashes were overpredicted over the western Pacific. Over the US the mass flux approach also performed better than the convective precipitation scheme (see Fig. 26.1).

$$F_{CG} = -0.234 + 0.308M - 0.719M^2 + 0.523M^3 - 0.0371M^4$$

Grewe et al. (2001) introduced a combination of updraft velocity w (m s^{-1}) and cloud thickness D (m) in a prediction formula for the total flash rate F (flashes per minute).

$$F = 1.54 \times 10^{-5}(wD^{0.5})^{4.9}$$

The grid cell average updraft velocity had to be derived from the parameterized upward mass flux as follows:

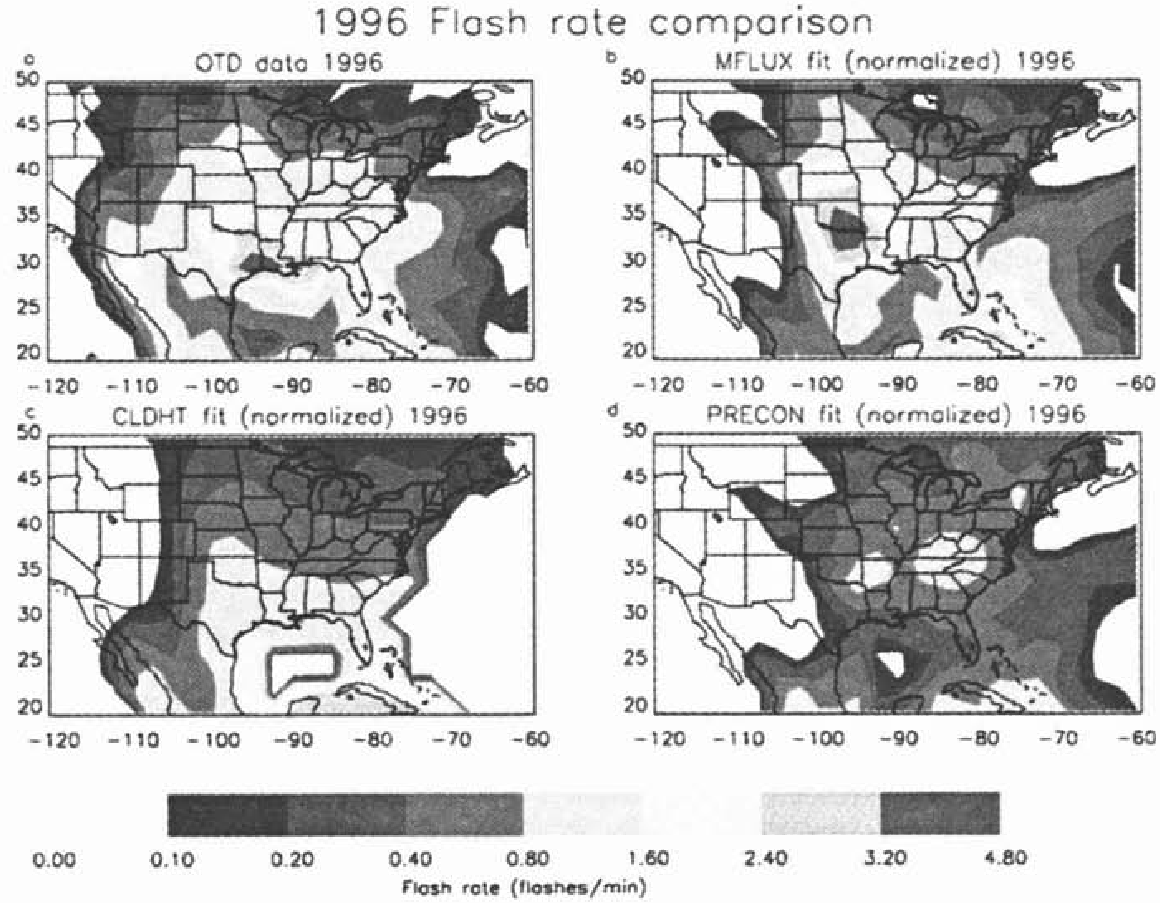

Fig. 26.1 Observed and predicted flash rates from three schemes over the US and Western Atlantic (from Allen and Pickering 2002) (See also Plate 49 in the Color Plate Section on page 629)

$$w = \Sigma(mf_i/\rho_i)(h_i/D)$$

where mf_i is the upward convective mass flux in kg m^{-2} s^{-1} in layer i, ρ_i is the mass density (kg m^{-3}), h_i is the thickness of the cloud layer i in meters, and D is the overall cloud thickness in meters.

Thermodynamic indicators of atmospheric instability such as convectively available potential energy (CAPE) may also be related to flash rate. Choi et al. (2005) developed such a relationship for use in a regional CTM.

Recently data from the TRMM satellite have been used to derive relationships between microphysical variables and lightning flash rates. Besides LIS, TRMM carries a precipitation radar. Data have been analyzed from these two instruments to derive two formulations which may be applicable in global models. Petersen et al. (2005) demonstrated a regime-independent relationship (see Fig. 26.2) between the column integrated precipitation ice mass, expressed as ice water path (kg/m^2), and the flash rate density (flashes/(km^2 d)). This relationship was found to be invariant between land, ocean and coastal regimes (in contrast to rainfall) and therefore applicable to the global scale. Cecil et al. (2005) compared LIS flash rates and precipitation features defined as contiguous areas having at least 20-dBZ near-surface reflectivity *or* 85-GHz polarization corrected temperature (PCT) $\leq$250 K. They found that

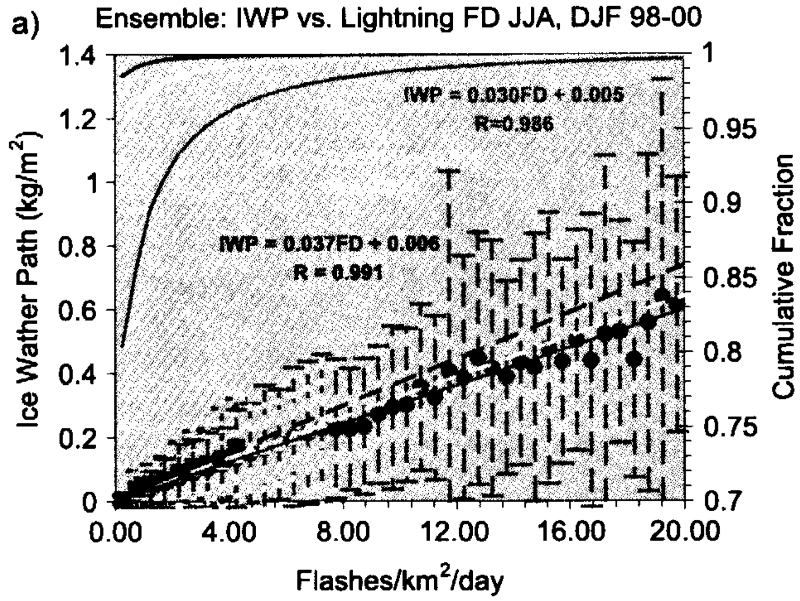

Fig. 26.2 Relationship of ice water path (IWP) vs. LIS flashes (from Petersen et al. 2005) (See also Plate 50 in the Color Plate Section on page 629)

for storms with the same brightness temperature, size and radar reflectivity aloft, the maritime storms are considerably less likely to produce lightning compared to continental storms. An explanation for these differences might be the lower cloud base of maritime clouds causing growth of large, precipitating raindrops below the freezing level, leaving less residual cloud water to become supercooled and form ice and graupel particles. Gauthier et al. (2006) and Sherwood et al. (2006) have further elucidated the roles of precipitation-size and cloud ice in determining lightning frequency. Recently, Futyan and DelGenio (2007) derived a relationship between flash rate (in flashes per minute per unit convective rain area of 300 km^{-2}) and the height (H in km) of the radar reflectivity top (defined as 17 dBZ). A slightly stronger relationship was found between flash rate and the depth (D_{cold} in km) of the layer between the freezing level and the radar reflectivity top (Fig. 26.3).

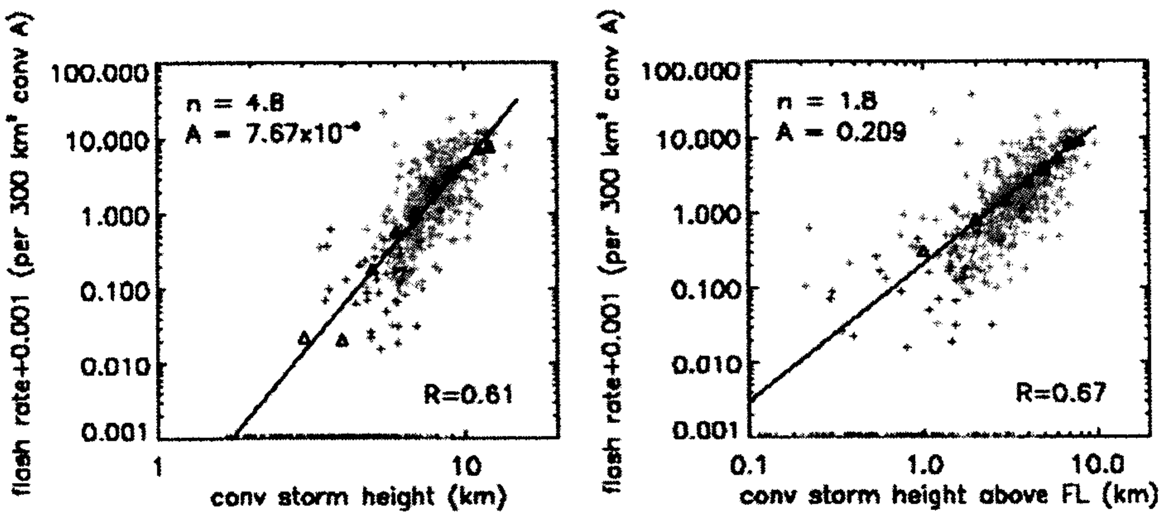

Fig. 26.3 Relationships of LIS flashes vs. radar top height (*left*) and vs. the depth between freezing level and radar top height (*right*) (from Futyan and DelGenio 2007) (See also Plate 51 in the Color Plate Section on page 630)

$$F = 7.67 \times 10^{-5} H^{4.8}$$
$$F = 0.209 D_{cold}^{1.8}$$

It was stated that this relationship captures both regional and continental-maritime contrasts in lightning occurrence and flash rate.

Treatment of cloud microphysics within atmospheric general circulation models has gradually become more sophisticated, and may soon reach the point where flash rate schemes using microphysical variables may be viable. Soon it may be possible to implement the parameterization suggested by Futyan and DelGenio (2007) in global models by using the predictions of the large particle top height. However, up to now the cloud microphysics included in the convection parameterization in the global models is too simplified to determine e.g. the ice water path, as discussed in Tost et al. (2007).

An additional microphysically-based scheme has been derived from ground-based radar and 3-D lightning mapping array data (Deierling et al. 2005, 2008). In this scheme, the flash rate is a function of the product of the upward flux of cloud ice crystals and of the downward flux of precipitation-size ice (graupel). It is unlikely that reliable estimates of such fluxes are going to be obtained from GCMs in the near future. However, this scheme has been employed in a cloud-resolving model by Barthe and Barth (2008). Dual-doppler radar and VHF total lightning observations have been examined by Deierling and Petersen (2008) to determine that the flash rate was well correlated with updraft volume above the $-5°C$ level when the vertical velocities exceeded 5 m/s in the Southeast US and 10 m/s over the High Plains of the US.

Recently, a new technique has been developed to obtain better geographic and temporal distribution of flashes than can be provided by any of the above described methods. This method (Sauvage et al. 2007; Martin et al. 2007) involves adjusting the flash rates obtained from the prediction equations using the OTD/LIS climatology. The adjustments can be performed on a regional and monthly basis. This procedure is also being employed in NASA's Global Modeling Initiative CTM (Allen et al. 2008).

26.3 NO Production per Flash

Estimation of NO production per flash has been performed using a variety of methods. The earliest estimates came from theoretical considerations of energy dissipation per flash and NO production per unit of energy. More recently, production estimates have been provided from laboratory spark measurements, detailed analysis of aircraft NO measurements taken in storms, cloud-resolved modeling constrained by observed flash rates and aircraft NO measurements from storm anvils, satellite NO_2 observations in relation to observed flashes, and best-fit calculations between observations and global model NO production rates. Details of each of these estimation techniques are presented below and in Section 26.5.

Price et al. (1997) formulated a lightning NO production scenario of 1100 moles per CG flash and 110 moles per IC flash through an analysis of theoretical concepts and observational data (1 mole $= 6.02 \times 10^{23}$ molecules $= 14$ g of nitrogen). Their analysis yielded 6.7 GJ per CG flash, and based on acoustical data an estimate of 10% of this value for IC flashes was made. An NO molecule production value of 10 $\times$ 10^{16} J^{-1} was assumed in estimating the moles per flash, which is in accordance with the average value of 8.5 $\pm$ 4.7 $\times$ 10^{16} estimated by Lawrence et al. (1995) from summarizing theoretical values found in the literature. However, the energy per CG flash value used by Price et al. is considerably greater than the 0.4 GJ per flash from the often cited Borucki and Chameides (1984) paper.

Laboratory spark experiments were conducted by Wang et al. (1998). They found that NO production increased nearly linearly with atmospheric pressure and quadratically with peak current of the flash. At surface pressure and typical values of peak current in the range 10–30 kA, the NO production was 15–40 $\times$ 10^{16} J^{-1}. These values are larger than that assumed by Price et al. (1997). However, Wang et al. estimated that ~50–150 moles per flash was produced by typical strokes (36 kA), which is well within the range of the average value of ~110$\pm$170 moles per flash estimated by Lawrence et al. (1995) from summarizing theoretical and laboratory values found in the literature and with the Price et al. (1997) value for IC flashes.

Aircraft measurements of NO in thunderstorm anvils conducted in field campaigns over the last 10–15 years (see Fig. 26.4) have yielded considerable information on NO production per flash, particularly when combined with observed lightning flash counts. Spikes in NO measured in the anvils of storms from the Stratosphere Troposphere Experiment: Radiation, Aerosol, Ozone (STERAO) field project in Colorado, USA in 1996 were analyzed by Stith et al. (1999) to yield a production rate in terms of NO molecules per unit flash length, which was extrapolated to flash lengths from 5 to 50 km. The resulting estimates ranges from 21

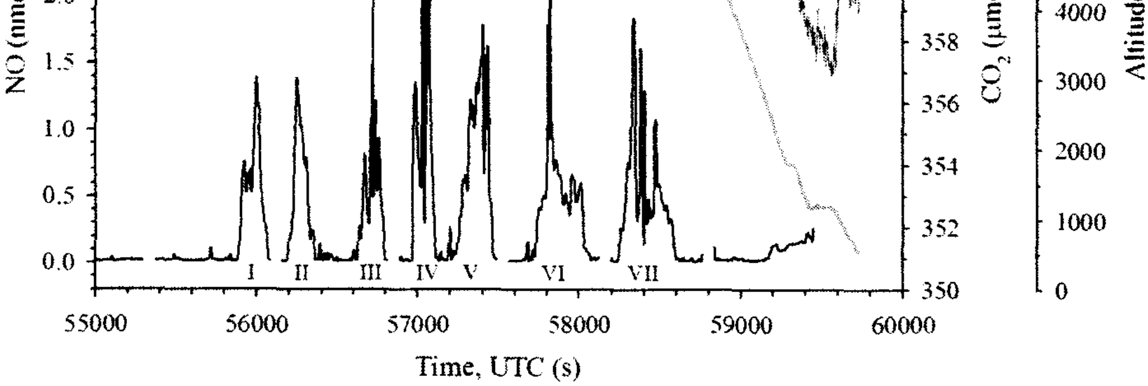

Fig. 26.4 Mixing ratio of NO (*black line*) and CO_2 (*dark gray line*), and altitude (*light gray line*) versus time from the EULINOX flight of 17 July 1998 over southern Germany. Seven penetrations of a squall line at different altitudes are labeled I–VII (from Huntrieser et al. 2002)

to 210 moles NO per flash, which is in accordance to the range found by Höller et al. (1999) and Huntrieser et al. (2002) during the airborne Lightning-produced NO_X (LINOX) and The European Lightning Nitrogen Oxides Project (EULINOX) experiments in 1996 and 1998 over Central Europe. Ridley et al. (2004) analyzed NO observations from the upper portion of anvils of Florida thunderstorms observed during the Cirrus Regional Study of Tropical Anvils and Cirrus Layers - Florida Area Cirrus Experiment (CRYSTAL-FACE) field project in 2002. These NO observations were related to observed flashes from the National Lightning Detection Network (NLDN) with a finding that the Florida storms analyzed had larger NO production rates per flash (55–382 moles) than storms from previous midlatitude field projects. Recently, Huntrieser et al. (2008) performed a detailed examination of the combination of anvil observations by aircraft and lightning strokes observed by the Lightning Location Network (LINET) system in Sao Paulo State in Brazil during the TROCCINOX experiments in 2004 and 2005. Results for tropical and subtropical thunderstorm events were contrasted in terms of NO production and stroke peak current, stroke release height and stroke component length. Simultaneous observations of LINET strokes and LIS flashes for one occasion were used to scale the estimated NO production rate per LINET stroke to the rate per LIS flash. The importance of scaling observed strokes or flashes from a local or regional system to global observations (OTD/LIS) was stressed, since observed production rates in single storms have been used to estimate the global LNO_X production rate. On average, tropical thunderstorms were found to produce less NO per LIS flash (86 moles) compared to subtropical thunderstorms (160 moles). The equivalent mean annual global LNO_X production rate was estimated to be 1.6 and 3.1 Tg/a, respectively. No distinct differences in the peak current frequency distribution were observed which could cause this difference in production rates. Also the difference in stroke release height was found to have only a minor influence. Instead the differences seem to be related to longer stroke component lengths in the subtropical thunderstorms compared to the tropical thunderstorms (factor of 2 difference). In fact, a larger mean flash length is supported by observations with a local flash detection systems (Huntrieser et al. 2008). The results suggest that the higher vertical wind shear observed in the subtropical compared to tropical thunderstorms during TROCCINOX may be responsible for the longer stroke component lengths. Up to now the flash length and the vertical wind shear has not been considered in any models simulating LNO_X.

Cloud-resolved chemical models have been used along with lightning flash rate and anvil NO observations to constrain estimates of NO production per flash. A series of cloud-resolved chemistry simulations described below has been conducted for individual storms observed in CRYSTAL-FACE (Ridley et al. 2004; Lopez et al. 2006), EULINOX (Huntrieser et al. 2002), and STERAO (Dye et al. 2000). All of these storms included research aircraft measuring chemical and meteorological properties at anvil levels. The time, location, and sometimes peak current of CG lightning occurrences were recorded by ground-based systems, and during STERAO and EULINOX, total lightning activity (IC + CG) was in addition mapped by a very high frequency (VHF) interferometer. Furthermore, all experiments included extensive satellite and radar observations of storm development. The dynamical evolution of each storm was simulated using a cloud-resolving model and the temperature, wind, and hydrometeor fields were then used to drive an offline cloud-scale chemical transport model. For each storm, various LNO_X production per flash scenarios were simulated and model results were compared with in-cloud aircraft observations of NO_X through use of mean profiles, column NO_X mass, and probability distribution functions to determine which scenario was the most appropriate.

A detailed description of the cloud-scale CTM developed at the University of Maryland is found in DeCaria et al. (2005). In this version of the model, LNO_X production is parameterized using observed IC and CG flash rates and a specified scenario of P_{IC} and P_{CG} to calculate the mass of NO injected into the cloud per time step. The NO produced by CG flashes is distributed unimodally in the vertical according to a Gaussian distribution centered at the altitude of the $-15^{\circ}C$ isotherm, while the NO produced by IC flashes is distributed bimodally with peaks at $-15^{\circ}C$ and at a cloud-height dependent anvil altitude. These distributions are based on the vertical distributions of VHF sources of IC and CG flashes presented in MacGorman and Rust (1998). A pressure dependence for NO production is also included. At each model level, the LNO_X is distributed uniformly to all grid cells within the 20 dBZ contour computed from simulated hydrometeor fields. An example of the NO_X distribution within the July 29, 2002 CRYSTAL-FACE storm is shown in Fig. 26.5.

Table 26.1 shows the production scenarios (Ott et al., 2008) estimated for five midlatitude and subtropical storms from the STERAO, EULINOX, and

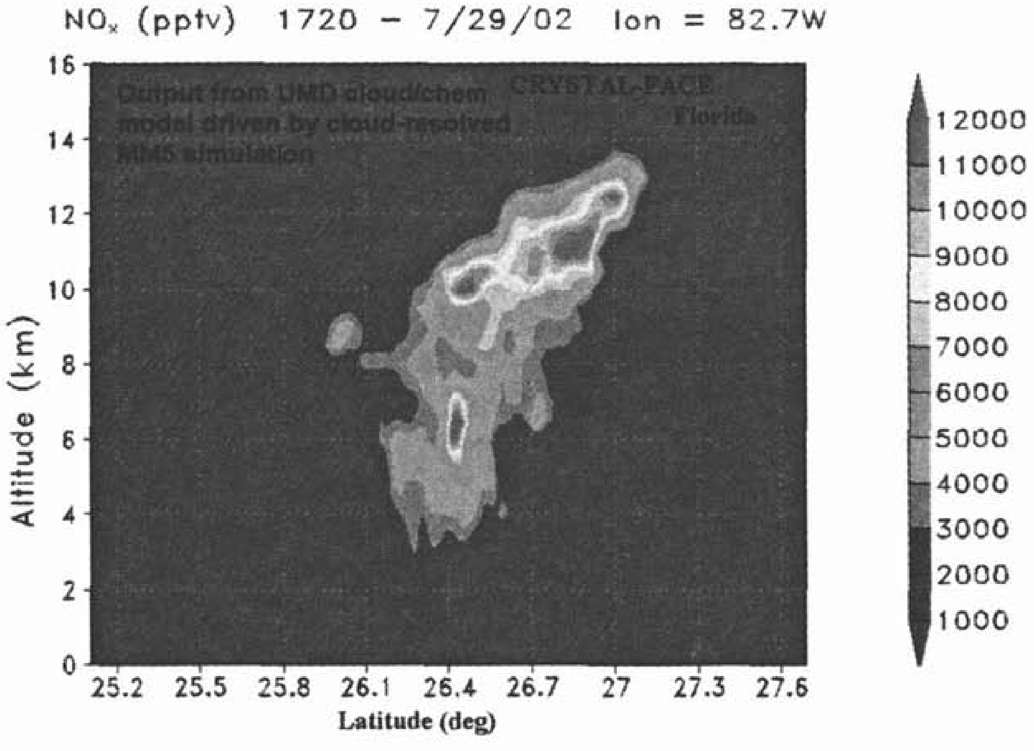

Fig. 26.5 Vertical cross section of NO_X ($pmol\ mol^{-1}$) through the July 29, 2002 CRYSTAL-FACE storm simulation using the University of Maryland cloud-scale CTM (See also Plate 52 in the Color Plate Section on page 630)

Table 26.1 LNO_X production in five midlatitude and subtropical storms simulated with the University of Maryland cloud-scale CTM and compared with theoretical estimates

Field project	Date	P_{CG} (moles/flash)	P_{IC}/P_{CG}
STERAO	July 10, 1996	460	0.75–1.0
STERAO	July 12, 1996	390	0.6
EULINOX	July 21, 1998	360	1.15
CRYSTAL-FACE	July 16, 2002	700	0.9
CRYSTAL-FACE	July 29, 2002	590	0.6–1.5
Mean		500	0.94
Price et al. (1997)		1100	0.1
Based on NALDN peak current (Orville et al. 2002)		508	NA

CRYSTAL-FACE campaigns using the University of Maryland cloud-scale CTM. Also listed is the production scenario from Price et al. (1997) which was used in calculating the vertical profiles of LNO_X mass presented in Pickering et al. (1998). In all cases, P_{CG} was estimated to be less than the 1100 moles per CG flash given in Price et al. (1997). In addition, the ratio of P_{IC} to P_{CG} was greater than the commonly assumed value of 0.1 presented by Price et al. (1997). Over the five storms simulated, the average estimated P_{CG} was 500 moles NO and the average P_{IC}/P_{CG} ratio was 0.94, corresponding to 470 moles NO for P_{IC}. For individual storms the P_{CG} values ranged from 360 to 700 moles per flash. The median peak current (16.5 kA for negative CG flashes and 19.8 kA for positive CG flashes which account for 10.9% of the total) of the North American Lightning Detection Network (NALDN) presented in Orville et al. (2002) would correspond to a P_{CG} value of 508 moles NO when using the Price et al. (1997) relationship between peak current and energy dissipated. This value agrees well with our estimate of 500 moles NO per CG flash. Therefore, the cases we have simulated appear to be representative. Assuming the average production scenario over the five simulated midlatitude and subtropical storms , an average IC to CG ratio of 3 (Boccippio et al. 2001), and a global flash rate of 44 s^{-1} (Christian et al. 2003) yields a global LNO_X source of ~9 Tg/a. The value of 9 Tg/a falls within the range obtained by Price et al. (1997) using the cloud-top height flash distribution based on International Satellite Cloud Climatology Project (ISCCP) deep convective clouds and 1100 moles/flash and 110 moles/flash for CG and IC flashes, respectively. However, the global LNO_X production value of 9 Tg/a is distinctly higher than the values resulting from extrapolating production per flash findings from laboratory measurements (1–3 Tg/a) and airborne field campaigns described above (3 Tg/a based on Huntrieser et al. (2002) and 1.1–7.5 Tg/a based on Ridley et al. (2004)). If the hypothesis of Huntrieser et al. (2008) regarding tropical flashes being less productive of LNO_X than midlatitude and subtropical flashes holds true for tropical cloud simulations, the global estimate based on such simulations would certainly decrease.

Other cloud-resolved models with chemical tracers have also been used in estimating NO production per flash. For example, Skamarock et al. (2003) used a cloud resolving model (COMMAS) to estimate the flux of NO_X out of the anvil of a STERAO thunderstorm and compared the computed flux with that from aircraft measurements to determine the production per flash (43 moles NO per flash averaged over total flashes). This analysis used the observed flash rate from a VHF interferometer which in addition detected a number of short duration flashes for which it is unknown if they produce NO molecules. Barthe and Barth (2008) simulated the same STERAO storm using the WRF-AqChem model of Barth et al. (2007). However, this simulation was performed with the flash rate parameterization of Deierling et al. (2008), which yielded 121 moles per flash (averaged over total flashes) and a conclusion from sensitivity studies that the short-duration flashes made very little NO. In this study the location of the LNO_X source was filamentary instead of volumetric, as in most previous studies, which yield a LNO_X production rate in the lower range compared to other studies.

Fehr et al. (2004) used a cloud-resolved version of MM5 with a lightning placement scheme in a simulation of the same EULINOX storm as simulated by Ott et al. (2007) with the cloud-scale CTM developed at the University of Maryland. Roughly similar values of P_{CG} (335 moles per flash from Fehr et al. and 360 moles per flash from Ott et al.) were obtained from the two models, but Fehr et al. obtained a larger value (1.4) of the per flash IC/CG production ratio than Ott et al. (1.15). A high value of this ratio is also needed to explain the high variability of NO data obtained from aircraft flying thought the thunderstorm anvil. It has been suggested that the NO concentration signatures would be smoother than observed if the NO originated largely from CG flashes (Höller et al. 2000). Ridley et al. (2005) have recommended that global models be run with the assumption of comparable production of NO by both types of lightning discharges.

Cloud-resolved models with explicit cloud electrification (Zhang et al. 2003; Barthe et al. 2005) have also been used in estimating LNO_X production. Such models simulate the processes of hydrometeor charging, charge separation, and the lightning discharge. Zhang et al. simulated a short-lived storm, generating 18 flashes over a 38-minute simulation, which yielded 2.03×10^{22} molecules NO per meter of flash channel, which is in the range of observations from other storms. The Barthe et al. model was applied to the 10 July STERAO storm, producing peaks of ~4 nmol mol^{-1} in the anvil similar to the airborne observations. LNO_X production per flash values were not provided from these models.

Case studies and global distributions of enhancements of tropospheric column NO_2 observations from satellite have also been used in estimates of LNO_X production. Beirle et al. (2006) used GOME tropospheric column observations of the number of NO_2 molecules per unit area of up to 4×10^{15} cm^{-2} over the Gulf of Mexico along with NLDN flash observations, and a climatological IC/CG ratio to estimate that these CG flashes produced 90 (32–240) moles of NO per flash and 1.7 (0.6–4.7) Tg/a globally. Boersma et al. (2005) used the global GOME NO_2 data along with a global chemical transport model to estimate a global LNO_X source strength of 1.6–6.4 Tg/a. Using the OTD/LIS climatological estimate of 44 s^{-1} for the global mean flash rate, this source strength implies a production per flash of 82–328 moles NO per flash.

26.4 Vertical Distribution of LNO_X Emissions

Many early CTMs made the assumption that LNO_X emissions were equally distributed in the vertical. The common assumptions were a constant mass or a constant mixing ratio (pressure dependent) with altitude. However, as more aircraft observations of NO_X and O_3 become available in the 1990s, it was noted that the influence of deep convection on LNO_X was most dominant in the middle to upper troposphere (e.g. Ridley et al. 1996; Huntrieser et al. 1998). Pickering et al. (1998) performed 2-D cloud-resolved model simulations of observed thunderstorms from several environments (midlatitude continental, tropical marine, and tropical continental) which were constrained with measured flash rates or observed anvil-NO_X mixing ratios These simulations were conducted with the Price et al. (1997) assumptions concerning LNO_X production per flash and with a flash placement scheme that was more crude than in more recent cloud-resolved modeling. The LNO_X mass in the model was integrated across the domain upon dissipation of the simulated storm and the percentage of this mass in each 1-km layer was computed. Profiles of the LNO_X mass derived in this way have been used in numerous global CTMs as the effective vertical distribution of LNO_X emissions (summarized in Schumann and Huntrieser 2007). The Pickering et al. (1998) profiles indicated that a majority of LNO_X is deposited above 8 km. A secondary maximum was found in the boundary layer as a result of downdrafts, yielding a C-shaped profile. Cloud-resolved simulations of a midlatitude supercell by Fehr et al. (2004) show even more pronounced maxima in the outflow region and boundary layer compared to Pickering et al. (1998). In an alternative scheme for simulations with a GCM, Kurz and Grewe (2002) used a simple quadratic parabola C-shape fit representing the LNO_X mass emissions.

However, more recent observations from STERAO, EULINOX and TROCCINOX have failed to find any significant LNO_X maxima in the boundary layer. More recent modeling studies (e.g., DeCaria et al. 2005; Ott et al. 2007), using more realistic flash placement schemes (e.g., the unimodal CG and bimodal IC flash channel distributions described above) and production per flash schemes, also do not show significant amounts of LNO_X in the boundary layer. Ott et al. (2008) summarized the results of 3-D cloud-resolved modeling of several midlatitude and subtropical storms (including those discussed by DeCaria et al. 2005; Ott et al. 2007) in terms of the vertical profile of LNO_X upon storm dissipation. Results show only small amounts ($<7\%$ in midlatitudes and $<3\%$ in subtropics) of LNO_X in the boundary layer, in accordance with observations. The other significant change from the Pickering et al. (1998) profiles is that the upper tropospheric maxima in percentage of LNO_X mass is now located at somewhat lower altitude in the new model output. In comparison to these prescribed vertical placements of LNO_X, a further approach has been developed by Mari et al. (2006) where a mass-flux formalism was implemented into a mesoscale model. No a-priori vertical placement of LNO_X is therefore necessary. The vertical distribution of LNO_X results from the redistribution of LNO_X emissions inside the convective scheme. Labrador et al. (2005) examined the effects of the vertical distribution of LNO_X emissions on atmospheric chemistry using a global CTM and noted that the shape of such vertical profiles is not only important in determining the large-scale distributions of NO_X but also of OH.

26.5 Applications and Performance of Global Models for Estimating the LNO_X Source and Its Impact on Global NO_X and Ozone Distributions

Global CTMs have been used to estimate the global source strength for LNO_X. This goal is achieved by comparing model mixing ratios of NO_X or O_3 with observations from ozonesondes, research aircraft, or satellite, followed by adjustment of the LNO_X source strength to obtain the best match with these observations. Numerous studies of this type have been conducted with a variety of models. Results of such analyses have been summarized by Schumann and Huntrieser (2007) and they suggest that the total global production is 2–8 Tg/a with a most likely value of 5 Tg/a. Early work used nitrate deposition data to constrain the LNO_X source strength (e.g., Penner et al. 1991). In more recent years sufficient aircraft and satellite data have become available to use for this purpose. Large data sets of aircraft observations influenced by LNO_X outflow from deep convection are valuable in allowing estimates of the global lightning source to be made with relative confidence. For example, Staudt et al. (2002, 2003) used data from the PEM Tropics A and B experiments to constrain the GEOS-Chem model and obtained estimates of 6 and 5 Tg/a, respectively. Satellite observations of tropospheric column NO_2 provide global coverage, but the NO_2 signal from lightning in standard satellite retrievals is only marginally greater than the noise in the measurements. Boersma et al. (2005) analyzed GOME NO_2 data in conjunction with the TM3-CTM and found that the global LNO_X source strength was between 1.6 and 6.4 Tg/a. Martin et al. (2007) used SCIAMACHY tropospheric column NO_2, which has better spatial coverage and resolution than GOME, in conjunction with the GEOS-Chem model (see Fig. 26.6). The resulting best-fit LNO_X source strength was 6 Tg/a. Schumann and Huntrieser (2007) report that simulations from several global models, with their preferred LNO_X parameterizations, were compared to trace gas observations from TROCCINOX (e.g. NO_X, CO and O_3). The evaluation method used (developed by U. Schumann) allows one to determine also the uncertainty range of the best estimate (based on the root-mean-square deviation between model results and observations) and the model sensitivity to the global LNO_X source rate. The best result was achieved with the TM4-CTM and a convective precipitation based parameterization which yield 4.8 $\pm$ 2.5 Tg/a for the annual global LNO_X production rate.

Tost et al. (2007) performed a comprehensive study with a global GCM where several state-of-the-art convection parameterizations were combined with different lightning parameterizations based on cloud-top height, updraft velocity and mass flux, and convective precipitation. A large variability in the results was found and none of the combinations approximately reproduced the observed lightning distributions from OTD/LIS. The updraft scheme developed by Grewe et al. (2001) in

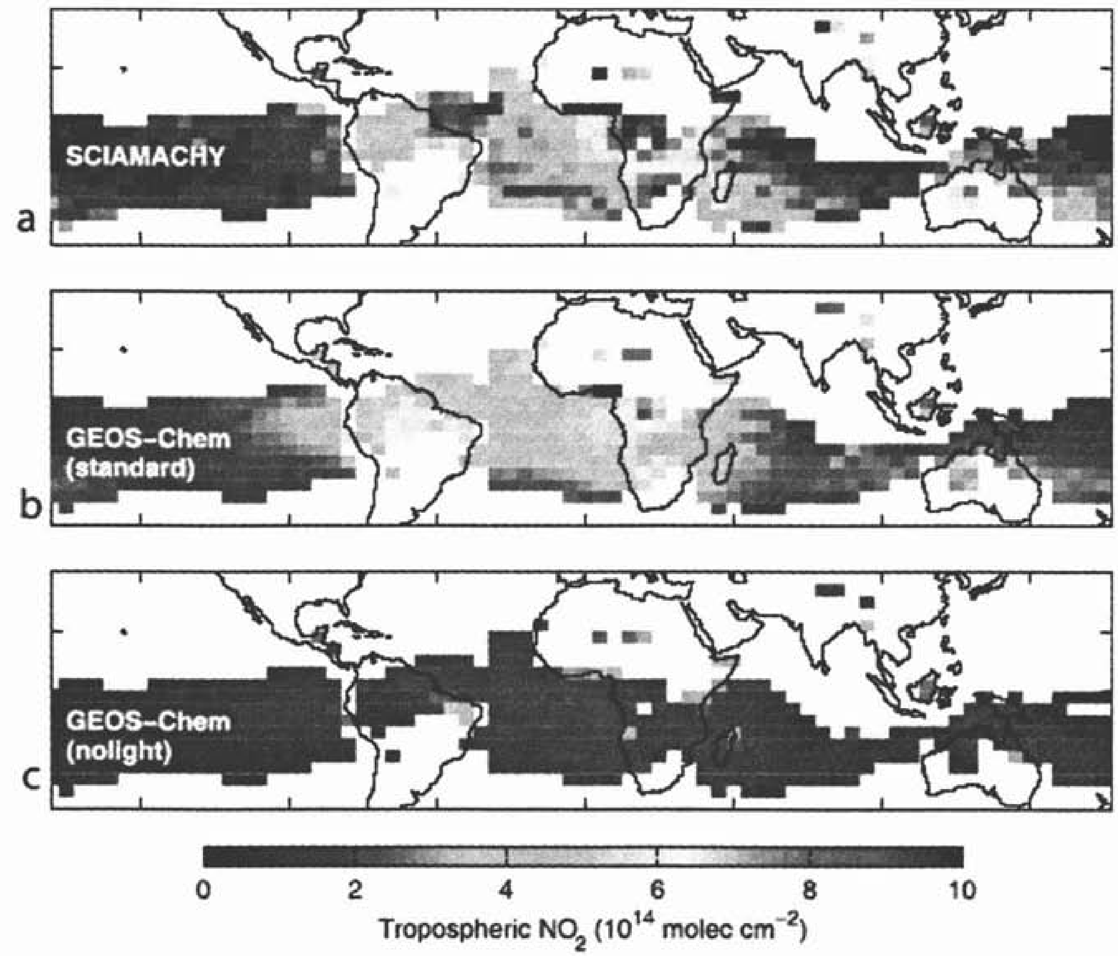

Fig. 26.6 SCIAMACHY tropospheric NO_2 column map compared with GEOS-Chem with 6 Tg/a from lightning and a GEOS-Chem run without lightning from Martin et al. (2007). Data are shown for regions where the model indicates the lightning contribution to be greater than 60% and the surface source contribution less than 25% (See also Plate 53 in the Color Plate Section on page 631)

combination with the Tiedtke (1989) scheme, which was especially developed for this GCM, scored better than most other combinations. However, the flash density over tropical oceans was overestimated and the maximum in flash density over Africa was underestimated. Even if the cloud-top height is not directly linked to cloud electrification, this approach was robust in combination with different convection parameterizations and with respect to both spatial and temporal variations of flash density. In contrast, Schumann and Huntrieser (2007) summarized that the LNO_X parameterizations based on the updraft and the convective precipitation schemes seem to simulate the variability of convection and lightning flash rates better than the parameterization based on cloud-top height.

Once a best-fit global LNO_X production is determined for a particular model, the model can be used to estimate the overall influence of LNO_X emissions on the large-scale distributions of NO_X and tropospheric ozone. Estimates of the fraction of NO_X due to LNO_X are generally obtained by running the model with and without the lightning source and subtracting the resulting NO_X fields. A general conclusion is that lightning is the dominant NO source for the upper troposphere over the entire year in the tropics and in the summer in the midlatitudes (e.g., Lamarque et al. 1996;

Berntsen and Isaksen 1999; Levy et al. 1999; Hauglustaine et al. 2001; Bond et al. 2002). Model simulations by Grewe (2007) show up to 70% of NOy (NOy includes all reactive odd nitrogen, also called fixed nitrogen) in the tropical upper troposphere is from lightning (see Fig. 17 in Schumann and Huntrieser (2007)). The impact of lightning on tropospheric reactive nitrogen NO_X and nitrogen reservoir species (HNO_3, peroxyacetyl nitrate (PAN), N_2O_5, and HNO_4) has been evaluated using a global chemical/transport model by Tie et al. (2001). Over 60% of the increase in total nitrogen species concentration when lightning was added to model was in the form of HNO_3. The increase in PAN accounts for approximately 20–30% of the nitrogen enhancement by lightning in the middle troposphere. Over 30% of tropospheric O_3 is formed by enhanced photochemistry due to LNO_X in the tropics

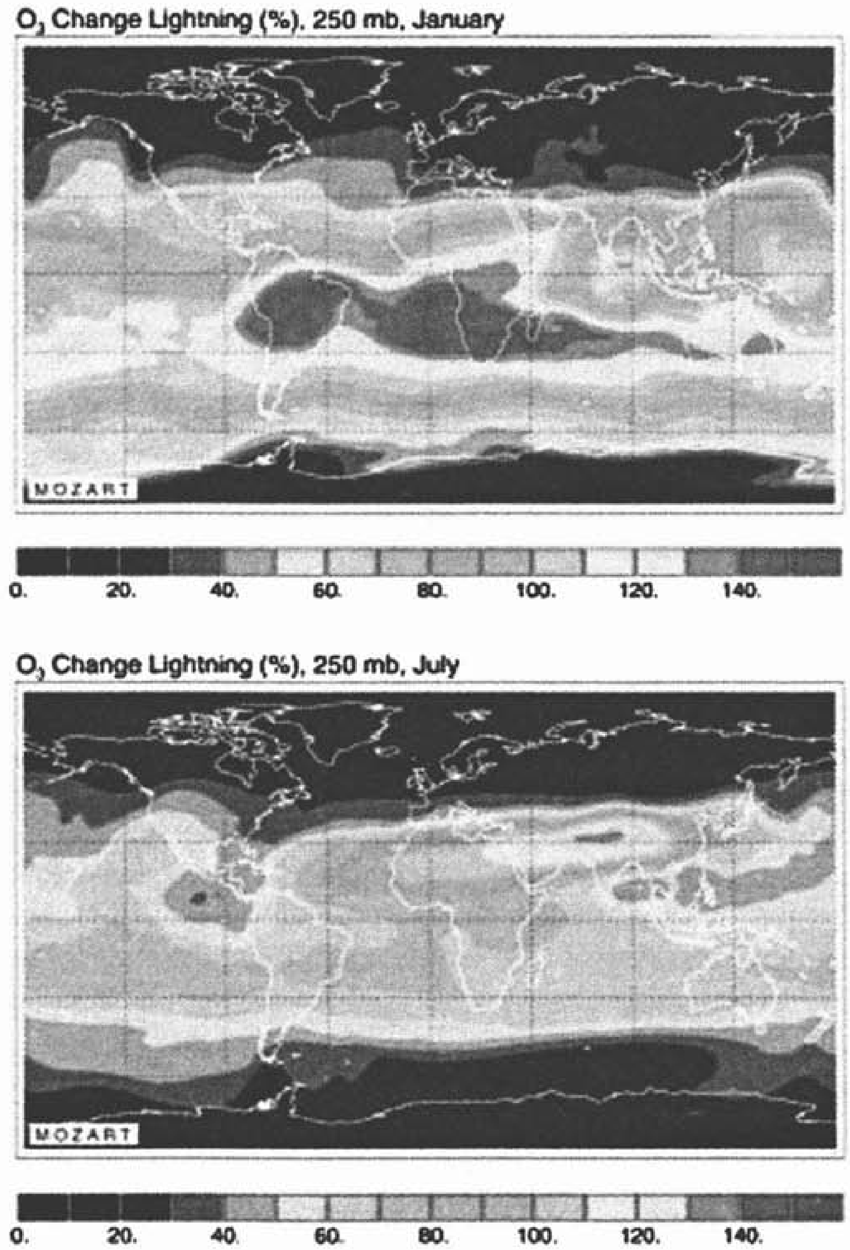

Fig. 26.7 Percentage increase in upper tropospheric (250 hPa) ozone due to lightning (from Hauglustaine et al. 2001) (See also Plate 54 in the Color Plate Section on page 632)

and over areas extending well into both hemispheres (Grewe 2007). Other models have yielded higher percentages. Hauglustaine et al. (2001) ran the MOZART CTM with and without LNO_X and found that ozone increases over the simulation without LNO_X were 150% over South America and Africa and >100% over the South Atlantic during the Southern Hemisphere summer. In Northern Hemisphere summer the ozone increases were 120% over South Asia and 20–50% over North America and Europe (see Fig. 26.7).

26.6 Conclusions

The range of values used for the global LNO_X production rate in more recent studies is still wide and range from 1 to 20 Tg/a, though more detailed recent studies support the smaller range of 2–8 Tg/a. However, for assessing the contribution to the NO_X and O_3 budgets especially in the tropics, the required LNO_X accuracy is about 1 Tg/a (Schumann and Huntrieser 2007). For most other tasks an accuracy of 5 Tg/a is enough, which is less than the range of best estimates suggested by Schumann and Huntrieser (2007). Considerably more research will be required to narrow the uncertainty to 1 Tg/a. Especially, more detailed field observations with 3-D lightning mapping arrays and airborne in-situ measurements of NO_X in the anvil outflow region of thunderstorms are required, which can help to improve the LNO_X parameterization in cloud-resolving models. These improvements can then be implemented to global models. Furthermore, detailed satellite measurements of lightning, precipitation and NO_2 with global cover are now available which are essential for evaluation of global models and can be used to adjust the models to the observations. The continuous increase in the resolution of satellite measurements and global models will also contribute to decrease the uncertainty range of LNO_X.

References

Allen, D., Pickering, K., Stenchikov, G., Thompson, A., Kondo, Y.: A three-dimensional total odd nitrogen (NOy) simulation during SONEX using a stretched-grid chemical transport model. J. Geophys. Res. **105**, 3851–3876 (2000). doi:10.1029/1999JD901029

Allen, D.J., Pickering, K.E.: Evaluation of lightning flash rate parameterizations for use in a global chemical transport model. J. Geophys. Res. **107**, 4711 (2002). doi:10.1029/2002JD002066

Allen, D.J., Pickering, K.E., Rodriguez, J.M, Duncan, B., Strahan, S., Logan, J., Damon, M.: Impact of lightning NO production on upper tropospheric NO_X and O_3 in the GMI model. J. Geophys. Res. in preparation (2008)

Barth, M.C., Kim, S.-W., Wang, C., et al.: Cloud-scale model intercomparison of chemical constituent transport in deep convection. Atmos. Chem. Phys. **7**, 4709–4731 (2007)

Barthe, C., Molinié, G., Pinty, J.-P.: Description and first results of an explicit electrical scheme in a 3D cloud resolving model. Atmos. Res. **76**, 95–113 (2005)

Barthe, C., Barth, M.C.: Evaluation of a new lightning-produced NO_X parameterization for cloud resolving models and its associated uncertainties. Atmos. Chem. Phys. Discuss. **8**, 6603–6651 (2008)

Beirle, S., Spichtinger, N., Stohl, A., et al.: Estimating the NO_X produced by lightning from GOME and NLDN data: a case study in the Gulf of Mexico. Atmos. Chem. Phys. **6**, 1075–1089 (2006)

Berntsen, T.K., Isaksen, I.S.A.: Effects of lightning and convection on changes in tropospheric ozone due to NO_X emissions from aircraft. Tellus **51B**, 766–788 (1999)

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

Boccippio, D.J.: Lightning scaling relations revisited. J. Atmos. Sci. **59**, 1086–1104 (2002)

Boersma, K.F., Eskes, H.J., Meijer, E.W., Kelder, H.M.: Estimates of lightning NO_X production from GOME satellite observations. Atmos. Chem. Phys. **5**, 2311–2331 (2005)

Bond, D.W., Steiger, S., Zhang, R., Tie, X., Orville, R.E.: The importance of NO_X production by lightning in the tropics. Atmos. Environ. **36**, 1509–1519 (2002)

Borucki, W.J., Chameides, W.L.: Lightning: estimates of the rates of energy dissipation and nitrogen fixation. Rev. Geophys. Space Phys. **22**, 363–372 (1984)

Cecil, D.J., Goodman, S.J., Boccippio, D.J., Zipser, E.J., Nesbitt, S.W.: Three years of TRMM precipitation features, Part I: radar, radiometric, and lightning characteristics. Mon. Wea. Rev. **133**, 543–566 (2005)

Choi, Y., Wang, Y., Zeng, T., Martin, R.V., Kurosu, T.P., Chance, K.: Evidence of lightning NO_X and convective transport of pollutants in satellite observations over North America. Geophys. Res. Lett. **32**, L02805 (2005). doi:10.1029/2004GL021436

Christian, H.J., Blakeslee, R.J., Boccippio, D.J., et al.: Global frequency and distribution of lightning as observed from space by the Optical Transient Detector. J. Geophys. Res. **108**, 4005 (2003). doi:10.1029/2002JD002347

DeCaria, A.J., Pickering, K.E., Stenchikov, G.L., Ott, L.E.: Lightning-generated NO_X and its impact on tropospheric ozone production: a three-dimensional modeling study of a Stratosphere-Troposphere Experiment: Radiation, Aerosols and Ozone (STERAO-A) thunderstorm. J. Geophys. Res. **110**, 1–13 (2005). doi:10.1029/2004JD005556

Deierling, W., Latham, J., Petersen, W.A., Ellis, S.M., Christian Jr., H.J.: On the relationship of thunderstorm ice hydrometeor characteristics and total lightning measurements. Atmos. Res. **76**, 114–126 (2005)

Deierling, W., Petersen, W., Latham, J., Ellis, S., Christian Jr., H.J.: The relationship between lightning activity and ice fluxes in thunderstorms. J. Geophys. Res. 113, D15210 (2008), doi: 10.1029/2007JD009700

Deierling, W., Petersen, W.: Total lightning activity as an indicator of updraft characteristics. J. Geophys. 113, D16210 (2008), doi: 10.1029/2007JD009598

Dye, J.E., Ridley, B.A., Skamarock, W., et al.: An overview of the Stratospheric-Tropospheric Experiment: Radiation, Aerosols, and Ozone (STERAO)-Deep Convection experiment with results for the July 10, 1996 storm. J. Geophys. Res. **105**, 10023–10045 (2000)

Fehr, T., Höller, H., Huntrieser, H.: Model study on production and transport of lightning-produced NO_X in a EULINOX supercell storm. J. Geophys. Res. **109**, 1–17 (2004). doi:10.1029/2003JD003935

Futyan, J.M., Del Genio, A.D.: Relationships between lightning and properties of convective cloud clusters. Geophys. Res. Lett. **34**, L15705 (2007). doi:10.1029/2007GL030227

Gauthier, M.L., Petersen, W.A., Carey, L.D., Christian Jr., H.J.: Relationship between cloud-to-ground lightning and precipitation ice mass: a radar study over Houston. Geophys. Res, Lett. **33**, L20803 (2006). doi:10.1029/2006GL027244

Grewe, V., Brunner, D., Dameris, M., Grenfell, J.L., Hein, R., Shindell, D., Staehelin, J.: Origin and variability of upper tropospheric nitrogen oxides and ozone at northern mid-latitudes. Atmos. Environ. **35**, 3421–3433 (2001)

Grewe, V.: Impact of climate variability on tropospheric ozone. Sci. Total Environ. **374**, 167–181 (2007). doi:10.1016/j.scitotenv.2007.01.032

Hauglustaine, D., Emmons, L., Newchurch, M., Brasseur, G., Takao, T., Matsubara, K., Johnson, J., Ridley, B., Stith, J., Dye, J.: On the role of lightning NO_X in the formation of tropospheric ozone plumes: a global model perspective. J. Atmos. Chem. **38**, 277–294 (2001)

Höller, H., Finke, U., Huntrieser, H., Hagen, M., Feigl, C.: Lightning-produced NO_X (LINOX): experimental design and case study results. J. Geophys. Res. **104**, 13911–13922 (1999). doi:10.1029/1999JD900019

Höller, H., Fehr, T., Thery, C., Seltmann, J., Huntrieser, H.: Radar, lightning, airborne observations and modelling of a supercell storm during EULINOX. Phys. Chem. Earth B, **25**, 1281–1284 (2000)

Huntrieser, H., Schlager, H., Feigl, C., Höller, H.: Transport and production of NO_X in electrified thunderstorms: survey of previous studies and new observations at midlatitudes. J. Geophys. Res. **103**, 28247–28264 (1998). doi:10.1029/98JD02353

Huntrieser, H., Feigl, C., Schlager, H., et al.: Airborne measurements of NO_X, tracer species, and small particles during the European lightning nitrogen oxides experiment. J. Geophys. Res. **107**, 4113 (2002). doi:10.1029/2000JD000209

Huntrieser, H., Schumann, U., Schlager, H., Höller, H., Giez, A., Betz, H.-D., Brunner, D., Forster, C., Pinto Jr., O., Calheiros, R.: Lightning activity in Brazilian thunderstorms during TROCCINOX: implications for NO_X production. Atmos. Chem. Phys. **8**, 921–953 (2008)

Intergovernmental Panel on Climate Change: Climate Change 2007 – The Physical Science Basis, Contribution of Working group 1 to the Fourth Assessment Report, 996 pp. Cambridge University Press, Cambridge (2007)

Kurz, C., Grewe, V.: Lightning and thunderstorms, Part I: Observational data and model results. Meteorol. Z. **11**, 379–393 (2002)

Labrador, L.J., von Kuhlmann, R., Lawrence, M.G.: The effects of lightning-produced NO_X and its vertical distribution on atmospheric chemistry: sensitivity simulations with MATCH-MPIC. Atmos. Chem. Phys. **5**, 1815–1834 (2005)

Lamarque, J.F., Brasseur, G.P., Hess, P.G., Mueller, J.F.: Three-dimensional study of the relative contributions of the different nitrogen sources in the troposphere. J. Geophys. Res. **101**, 22955–22968 (1996)

Lawrence, M.G., Chameides, W.L., Kasibhatla, P.S., Levy II, H., Moxim, W.: Lightning and atmospheric chemistry: The rate of atmospheric NO production, in: Volland, H. (ed.) Handbook of Atmospheric Electrodynamics, pp. 189–202. CRC Press, Boca Raton, Florida (1995)

Levy II, H., Moxim, W.J., Klonecki, A.A., Kasibhatla, P.S.: Simulated tropospheric NO_X: its evaluation, global distribution and individual source contributions. J. Geophys. Res. **104**, 26279–26306 (1999)

Lopez, J.P., Fridlind, A.M., Jost, H.-J., et al.: CO signatures in subtropical convective clouds and anvils during CRYSTAL-FACE: an analysis of convective transport and entrainment using observations and a cloud-resolving model. J. Geophys. Res. **111**, D09305 (2006). doi:10.1029/2005JD006104

MacGorman, D.R., Rust, W.D.: The Electrical Nature of Storms. 422 pp. Oxford University Press, Oxford (1998)

Mari, C., Mari, C., Chaboureau, J.P., Pinty, J.P., et al.: Regional lightning NO_X sources during the TROCCINOX experiment. Atmos. Chem. Phys. **6**, 5559–5572 (2006)

Martin, R.V., Sauvage, B., Folkins, I., Sioris, C.E., Boone, C., Bernath, P., Ziemke, J.: Space-based constraints on the production of nitric oxide by lightning. J. Geophys. Res. **112**, D09309 (2007). doi:10.1029/2006JD007831

Meijer, E.W., van Velthoven, P.F.J., Brunner, D.W., Huntrieser, H., Kelder, H.: Improvement and evaluation of the parameterisation of nitrogen oxide production by lightning. Phys. Chem. Earth. **26**, 577–583 (2001)

Orville, R.E., Huffines, G.R., Burrows, W.R., Holle, R.L., Cummins, K.L.: The North American lightning detection network (NALDN) – First results: 1998–2000. Mon. Wea. Rev. **130**, 2098–2109 (2002)

Ott, L.E., Pickering, K.E., Stenchhikov, G.L., Huntrieser, H., Schumann, U.: Effects of lightning NO_X production during the 21 July European lightning nitrogen oxides project storm studied with a three-dimensional cloud-scale chemical transport model. J. Geophys. Res. **112**, D05307 (2007). doi:10.1029/2006JD007365

Ott, L., Pickering, K.E., DeCaria, A., Stenchikov, G., Lin, R.-F., Wang, D., Lang, S., Tao, W.-K.: Production of lightning NO_X and its vertical distribution calculated from 3-D cloud-scale chemical transport model simulations. J. Geophys. Res. in preparation (2008)

Penner, J.E., Atherton, C.S., Dignon, J., Ghan, S.J., Walton, J.J., Hameed, S.: Tropospheric nitrogen: a three-dimensional study of sources, distributions, and deposition. J. Geophys. Res. **96**, 959–990 (1991). doi:10.1029/90JD02228

Petersen, W.A., Rutledge, S.A.: On the relationship between cloud-to-ground lightning and convective rainfall. J. Geophys. Res. **103**, 14025–14040 (1998). doi:10.1029/97JD02064

Petersen, W.A., Christian, H.J., Rutledge, S.A.: TRMM observations of the global relationship between ice water content and lightning. Geophys. Res. Lett. **32**, 1–4 (2005). doi:10.1029/2005GL023236

Pickering, K.E., Wang, Y., Tao, W.K., Price, C., Müller, J.F.: Vertical distributions of lightning NO_X for use in regional and global chemical transport models. J. Geophys. Res. **103**, 31203–31216 (1998). doi:10.1029/98JD02651

Price, C., Rind, D.: A simple lightning parameterization for calculating global lightning distributions. J. Geophys. Res. **97**, 9919–9933 (1992). doi:10.1029/92JD00719

Price, C., Rind, D.: What determines the cloud-to-ground lightning fraction in thunderstorms?. Geophys. Res. Lett. **20**, 463–466 (1993). doi:10.1029/93GL00226

Price, C., Penner, J., Prather, M.: NO_X from lightning, 1. Global distribution based on lightning physics. J. Geophys. Res. **102**, 5929–5941 (1997). doi:10.1029/96JD03504

Ridley, B.A., Dye, J.E., Walega, J.G., Zheng, J., Grahek, F.E., Rison, W.: On the production of active nitrogen by thunderstorms over New Mexico. J. Geophys. Res. **101**, 20985–21005 (1996)

Ridley, B., Ott, L., Pickering, K., et al.: Florida thunderstorms: a faucet of reactive nitrogen to the upper troposphere. J. Geophys. Res. **109**, 1–19 (2004). doi:10.1029/2004JD004769

Ridley, B.A., Pickering, K.E., Dye, J.E.: Comments on the parameterization of lightning-produced NO in global chemistry-transport models, Atmos. Environ., **39**, 6184–6187 (2005)

Sauvage, B., Martin, R.V., van Donkelaar, A., Liu, X., Chance, K., Jaeglé, L., Palmer, P.I., Wu, S., Fu, T.-M.: Remote sensed and in situ constraints on processes affecting tropical tropospheric ozone. Atmos. Chem. Phys. **7**, 815–838 (2007)

Schumann, U., Huntrieser, H.: The global lightning-induced nitrogen oxides source. Atmos. Chem. Phys. **7**, 3823-3907 (2007)

Sherwood, S.C., Phillips, V.T.J., Wettlaufer, J.S.: Small ice crystals and the climatology of lightning. Geophys. Res. Lett. **33**, L05804 (2006). doi:10.1029/2005GL025242

Skamarock, W.C., Dye, J.E., Defer, E., Barth, M.C., Stith, J.L., Ridley, B.A., Baumann, K.: Observational and modeling-based budget of lightning-produced NO_X in a continental thunderstorm. J. Geophys. Res. **108**, 4305 (2003). doi:10.1029/2002JD002163

Staudt, A.C., Jacob, D.J., Logan, J.A., Bachiochi, D., Krishnamurti, T.N., Poisson, N.: Global chemical model analysis of biomass burning and lightning influences over the South Pacific in austral spring. J. Geophys. Res. **107**, 4200 (2002). doi:10.1029/2000JD000296

Staudt, A.C., Jacob, D.J., Ravetta, F., Logan, J.A., Bachiochi, D., Krishnamurti, T.N., Sandholm, S., Ridley, B., Singh, H.B., Talbot, B.: Sources and chemistry of nitrogen oxides over the tropical Pacific. J. Geophys. Res. **108**, 8239 (2003). doi:10.1029/2002JD002139

Stith, J., Dye, J., Ridley, B., Laroche, P., Defer, E., Baumann, K., Huebler, G., Zerr, R., Venticinque, M.: NO signatures from lightning flashes. J. Geophys. Res. **104**, 16081–16089 (1999)

Tie, X., Zhang, R., Brasseur, G., Emmons, L., Lei, W.: Effects of lightning on reactive nitrogen and nitrogen reservoir species in the troposphere. J. Geophys. Res. **106**, 3167–3178 (2001). doi:10.1029/2000JD900565

Tiedtke, M.: A comprehensive mass flux scheme for cumulus parameterization in large-scale models. Mon. Wea. Rev. **117**, 1779–1800 (1989)

Tost, H., Jöckel, P., Lelieveld, J.: Lightning and convection parameterisations – uncertainties in global modeling. Atmos. Chem. Phys. **7**, 4553–4568 (2007)

Ushio, T., Heckman, S.J., Boccippio, D.J., Christian, H.J., Kawasaki, Z.-I.: A survey of thunderstorm flash rates compared to cloud top height using TRMM satellite data. J. Geophys. Res. **106**, 24089–24095 (2001)

Vonnegut, B.: Some facts and speculations concerning the origin and role of thunderstorm electricity, in: Atlas, D., Booker, D.R., Byers, H., et al. (eds.) Severe Local Storms, Meteorol. Monogr., vol. 5, no. 27., pp. 224–241. Am. Meteor. Soc., Boston (1963)
Wang, Y., DeSilva, A.W., Goldenbaum, G.C., Dickerson, R.R.: Nitric oxide production by simulated lightning: Dependence on current, energy, and pressure. J. Geophys. Res. **103**, 19149–19159 (1998)
Williams, E.: Large-scale charge separation in thunderclouds. J. Geophys. Res. **90**, 6013–6025 (1985)
Zel'dovich, Y.B., Raizer, Y.P.: Physics of Shock Waves and High Temperature Hydrodynamic Phenomena. 566–571 pp. Academic, San Diego, CA (1967)
Zhang, X., Helsdon Jr., J.H., Farley, R.D.: Numerical modelling of lightning-produced NO_X using an explicit lightning scheme: 2. Three-dimensional simulation and expanded chemistry. J. Geophys. Res. **108**, 4580 (2003). doi:10.1029/2002JD003225