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## Space-based constraints on the production of nitric oxide by lightning

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[1] We interpret observations of trace-gases from three satellite platforms to provide top-down constraints on the production of NO by lightning. The space-based observations are tropospheric NO<sub>2</sub> columns from SCIAMACHY, tropospheric O<sub>3</sub> columns from OMI and MLS, and upper tropospheric HNO<sub>3</sub> from ACE-FTS. A global chemical transport model (GEOS-Chem) is used to identify locations and time periods in which lightning would be expected to dominate the trace gas observations. The satellite observations are sampled at those locations and time periods. All three observations exhibit a maximum in the tropical Atlantic region and a minimum in the tropical Pacific. This wave-1 pattern is driven by injection of lightning NO into the upper troposphere over the tropical continents, followed by photochemical production of NO<sub>2</sub>, HNO<sub>3</sub>, and O<sub>3</sub> during transport. Lightning produces a broad enhancement over the tropical Atlantic and Africa of  $2\text{--}6 \times 10^{14}$  molecules NO<sub>2</sub> cm<sup>-2</sup>,  $4 \times 10^{17}$  molecules O<sub>3</sub> cm<sup>-2</sup> (15 Dobson Units), and 125 pptv of upper tropospheric HNO<sub>3</sub>. The lightning background is 25–50% weaker over the tropical Pacific. A global source of  $6 \pm 2$  Tg N yr<sup>-1</sup> from lightning in the model best represents the satellite observations of tropospheric NO<sub>2</sub>, O<sub>3</sub>, and HNO<sub>3</sub>.

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### 1. Introduction

[2] Nitrogen oxide radicals (NO<sub>x</sub> ≡ NO + NO<sub>2</sub>) originating from combustion, lightning, and soils largely control tropospheric O<sub>3</sub> production [Kasibhatla *et al.*, 1991; Penner *et al.*, 1991; Murphy *et al.*, 1993; Jacob *et al.*, 1996]. Lightning remains the most uncertain source of NO<sub>x</sub> [Lee *et al.*, 1997]. Most estimates of the global lightning NO<sub>x</sub> source have employed a bottom-up approach based on estimates of global flash rates and the number of molecules of NO produced per flash [Price *et al.*, 1997a; Labrador *et al.*, 2005]. Top-down constraints based on satellite observations of trace gases could reduce considerably this uncertainty.

[3] Table 1 contains recent estimates of global lightning NO<sub>x</sub> production. Global estimates vary by more than an order of magnitude over 1–20 Tg N yr<sup>-1</sup>. Flash rates explain only a small fraction of this uncertainty. Recent estimates of global flash rates derived from space-based

measurements from the Optical Transient Detector (OTD) are  $44 \pm 5$  flashes s<sup>-1</sup> [Christian *et al.*, 2003], 50–75% of earlier estimates over the last decade. The remaining uncertainty in global estimates from the bottom-up approach arises from the production of NO/flash, for which recent values vary by an order of magnitude, in part reflecting spatial variation in the frequency [Bocchippio *et al.*, 2001] and uncertainty in the relative yield [Ridley *et al.*, 2005] of cloud-ground (CG) and intracloud (IC) flashes.

[4] Levy *et al.* [1996] introduced an alternative top-down approach of comparing airborne observations of NO<sub>x</sub> and NO<sub>y</sub> with a global model simulation and found that this approach yielded a tight constraint on the lightning NO<sub>x</sub> source. Boersma *et al.* [2005] extended their method using satellite measurements of tropospheric NO<sub>2</sub> columns from the Global Ozone Monitoring Instrument (GOME). We develop the top-down approach here using global observations of tropospheric NO<sub>2</sub> columns from the Scanning Imaging Absorption Spectrometer for Atmospheric Characterization (SCIAMACHY) satellite instrument [Bovensmann *et al.*, 1999], tropospheric O<sub>3</sub> columns from the Ozone Monitoring Instrument (OMI) [Levelt *et al.*, 2006] and Microwave Limb Sounder (MLS) [Waters *et al.*, 2006] instruments, and upper tropospheric HNO<sub>3</sub> measurements from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) [Bernath *et al.*, 2005]. These multiple species provide three independent constraints on the lightning NO<sub>x</sub> source.

[5] A major challenge in interpreting satellite observations of a tropospheric column is ambiguity in the altitude of the gas. Surface sources and free tropospheric sources both

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**Table 1.** Recent Estimates of Global Lightning NO<sub>x</sub> Production

Study	Production Rate, Tg N yr <sup>-1</sup>	Approach	Flash, s <sup>-1</sup>	10 <sup>25</sup> atoms, N/flash
Levy et al. [1996]	2–6	Comparison of global model with airborne observations of NO <sub>x</sub> and NO <sub>y</sub>		
Ridley et al. [1996]	2–5	Extrapolation of airborne measurements of New Mexico storms	100	
Price et al. [1997a]	12 (5–20) <sup>a</sup>	Lightning physics	70–100	25 (4–105) <sup>b</sup>
Price et al. [1997b]	13 (5–25)	Constraints from atmospheric electricity		
Huntrieser et al. [1998]	4 (0.3–22)	Extrapolation of airborne measurements of storms in Europe		4–30
H. Wang et al. [1998]	3–8	Bottom-up from laboratory measurements	30–100	3
Y. Wang et al. [1998]	3	Comparison of global model with airborne observations in North Atlantic		
Nesbitt et al. [2000]	0.9	Global satellite flash counts	57	2 (1–6)
Bond et al. [2002]	6 (0.5–9)	LIS flash counts	45	20 (7–70)
Huntrieser et al. [2002]	3 (1–20)	Extrapolation of airborne measurements of storms in Europe	65	8
Martin et al. [2002b]	6	Comparison of global model with O <sub>3</sub> from TOMS and sondes		
Beirle et al. [2004]	3 (0.8–14)	Extrapolation of GOME NO <sub>2</sub> and LIS observations over Australia	63	6 (2–30)
Ridley et al. [2004]	1–8	Extrapolation of measurements of Florida thunderstorms	44	3–23
Boersma et al. [2005]	1–6	Analysis of GOME NO <sub>2</sub> observations in southern tropics		
Labrador et al. [2005]	<20	Comparison of global model with airborne observations of NO <sub>x</sub>		
Beirle et al. [2006]	2 (0.6–5)	Extrapolation of GOME measurements of NO <sub>2</sub> in the Gulf of Mexico	44	5 (2–14)
This work	6 (4–8)	Comparison of global model with satellite observations of NO <sub>2</sub> , O <sub>3</sub> , and HNO <sub>3</sub>		

<sup>a</sup>The reported estimate range is given in parentheses.

<sup>b</sup>Represents the weighted average for intercloud and cloud-ground flashes.

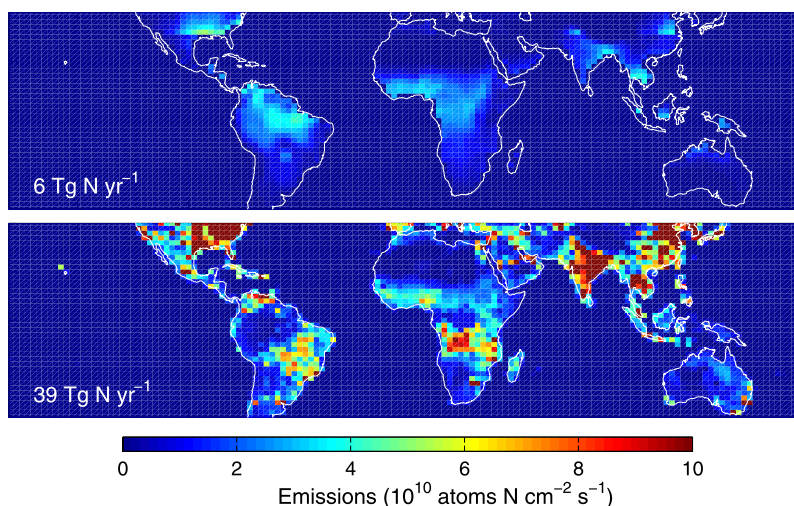
contribute to column enhancements. Figure 1 shows a current estimate of the distribution of NO<sub>x</sub> production by lightning and other sources. Most regions with a substantial lightning NO<sub>x</sub> source also contain considerable emissions from other sources, confounding a simple correlation between the column observation and lightning flashes. However, one major difference between the production of NO<sub>x</sub> from lightning and other sources is their altitude. Simulations with cloud resolving models suggest that most lightning NO<sub>x</sub> is deposited above 7 km [Pickering et al., 1998; Skamarock et al., 2003] where the lifetimes of NO<sub>x</sub>, HNO<sub>3</sub>, and O<sub>3</sub> are 5–10 times longer than in the lower troposphere. In the tropics, convection of NO<sub>x</sub> from surface sources is expected to make a minor contribution to upper tropospheric NO<sub>x</sub> compared with lightning NO<sub>x</sub> emissions [Levy et al., 1999]. Lightning NO<sub>x</sub> would be expected to influence a much broader region than surface sources [Lamarque et al., 1996]. An upper tropospheric air mass will be advected along a different trajectory than a lower tropospheric air-

mass that is influenced by surface emissions. Boersma et al. [2005] introduced the idea of a masking scheme to extract the lightning signal by minimizing the interference of surface NO<sub>x</sub> sources on satellite observations.

[6] In this work we use a global chemical transport model (GEOS-Chem) to identify the locations, regions, and species that are dominated by the effects of lightning NO<sub>x</sub>. Section 2 introduces the GEOS-Chem model. Section 3 compares the satellite observations of tropospheric NO<sub>2</sub>, O<sub>3</sub>, and HNO<sub>3</sub> with the model simulation in those regions. Conclusions are in section 4.

## 2. GEOS-Chem Model

[7] We use the GEOS-Chem global three-dimensional (3-D) chemical transport model to account for the photochemical evolution and transport of NO<sub>x</sub>. The simulation is driven by assimilated meteorological data from the Goddard Earth Observing System (GEOS-4) at the NASA Global



**Figure 1.** Annual mean NO<sub>x</sub> emissions used in the standard GEOS-Chem simulation from (top) lightning and (bottom) all other sources including fossil fuel combustion, biomass burning, biofuels, and soils.

Modeling and Assimilation Office (GMAO). We use here version 7-03-03 of GEOS-Chem ([www-as.harvard.edu/chemistry/trop/geos](http://www-as.harvard.edu/chemistry/trop/geos)) with additional midlatitude lightning NO<sub>x</sub> as described below. The GEOS-Chem model includes a detailed simulation of tropospheric ozone-NO<sub>x</sub>-hydrocarbon chemistry as well as of aerosols and their precursors. The ozone-NO<sub>x</sub>-hydrocarbon simulation was first described by *Bey et al.* [2001] with updates by *Fiore et al.* [2002] and *Martin et al.* [2002b, 2003b]. The model presently includes sulfate, nitrate, ammonium, black carbon, organic carbon, mineral dust, and sea salt [*Park et al.*, 2003; *Park et al.*, 2004; *Alexander et al.*, 2005; *Fairlie et al.*, 2006]. The aerosol and gaseous simulations are coupled through formation of sulfate and nitrate, HNO<sub>3</sub>(g)/NO<sub>3</sub><sup>-</sup> partitioning of total inorganic nitrate, heterogeneous chemistry on aerosols [*Jacob*, 2000; *Evans and Jacob*, 2005], and aerosol effects on photolysis rates [*Martin et al.*, 2003b].

[8] Figure 1 shows the annual global distribution of NO<sub>x</sub> emissions in GEOS-Chem for all sources. Anthropogenic NO<sub>x</sub> emissions are based on the Global Emission Inventory Activity (GEIA) [*Benkovitz et al.*, 1996] as described by *Bey et al.* [2001]. Soil NO<sub>x</sub> emissions are computed using a modified version of the algorithm of *Yienger and Levy* [1995] with the canopy reduction factors described by *Y. Wang et al.* [1998]. The biomass burning inventory is seasonally varying and is based on satellite observations of fires over 1996–2000 from the Along Track Scanning Radiometer (ATSR) as derived by *Duncan et al.* [2003]. Emissions of lightning NO<sub>x</sub> are linked to deep convection following the parameterization of *Price and Rind* [1992] with vertical profiles from *Pickering et al.* [1998] as implemented by *Y. Wang et al.* [1998]. The midlatitude lightning NO<sub>x</sub> source is increased here by a factor of four to 1.6 Tg N yr<sup>-1</sup> following *Hudman et al.* [2007] and *Martin et al.* [2006] to correct a model bias in simulated NO<sub>x</sub> versus INTEX-A aircraft observations, to be consistent with NO production per flash estimates from recent cloud-scale modeling [*DeCaria et al.*, 2005; *Pickering et al.*, 2006], and to account for recent observational evidence that midlatitude lightning may produce more NO per flash than tropical lightning [*Huntrieser et al.*, 2006]. The model chemical fields have been evaluated versus in situ and satellite observations throughout the tropics [*Bey et al.*, 2001; *Chandra et al.*, 2002; *Martin et al.*, 2002b; *Chandra et al.*, 2003; *Martin et al.*, 2003a; *Jaeglé et al.*, 2005; *Kim et al.*, 2005; *Folkens et al.*, 2006; *Liu et al.*, 2006; *Sauvage et al.*, 2007a].

[9] We conduct eight sensitivity simulations with variable lightning source strength, or with NO<sub>x</sub> emissions excluded from either biomass burning or soils to examine the effect of these sources. Sensitivity simulations are conducted over May 2004 to April 2005, following a 4-month spinup. Two additional sensitivity studies are used to assess the robustness of the conclusions to spatial variation in the lightning source. Estimates of the relative vertical distribution of lightning NO<sub>x</sub> is affected by uncertainty in the NO<sub>x</sub> production per flash for intracloud (P<sub>IC</sub>) and cloud-ground (P<sub>CG</sub>) flashes. The vertical profiles from *Pickering et al.* [1998] used in the standard simulation were determined for a P<sub>IC</sub>/P<sub>CG</sub> ratio of 0.1, based on the work of *Price et al.* [1997a]. There is evidence for a P<sub>IC</sub>/P<sub>CG</sub> ratio of 0.5–1 [*Gallardo and Cooray*, 1996; *DeCaria et al.*, 2000; *Zhang et al.*, 2003; *Fehr et al.*, 2004; *DeCaria et al.*, 2005; *Ott et*

*al.*, 2007]. One sensitivity study uses a P<sub>IC</sub>/P<sub>CG</sub> ratio of 0.75 in which the additional NO<sub>x</sub> from intracloud flashes is distributed within the cloud anvil.

[10] *Allen and Pickering* [2002] found that the *Price and Rind* [1992] lightning parameterization does not fully capture the observed distribution of lightning activity. We conduct another sensitivity study in which the spatial distribution of lightning NO<sub>x</sub> production is rescaled to match 10 years of seasonally varying observations of lightning flashes from the OTD and the Lightning Imaging Sensor (LIS) as implemented by *Sauvage et al.* [2007a].

### 3. Top-Down Constraints on Lightning NO<sub>x</sub> Emissions

[11] Satellite retrievals of tropospheric NO<sub>2</sub> columns, tropospheric O<sub>3</sub> columns, and upper tropospheric HNO<sub>3</sub> provide three independent constraints on the production of NO from lightning. We examine each species in turn.

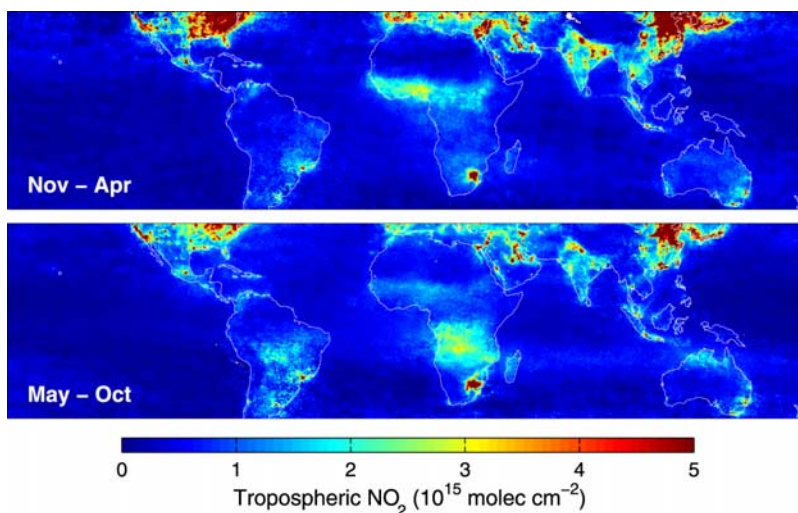
#### 3.1. Analysis of Tropospheric NO<sub>2</sub> Columns

[12] The SCIAMACHY instrument on board the ENVISAT satellite provides the capability for global retrieval of atmospheric NO<sub>2</sub> columns through observation of global backscatter [*Bovensmann et al.*, 1999]. The satellite was launched in March 2002 into a Sun-synchronous orbit, crossing the equator at about 1000 LT in the descending node. The SCIAMACHY instrument observes the atmosphere in the nadir view with a typical surface spatial resolution of 30 km along track by 60 km across track. Global coverage is achieved every 6 days.

[13] We use cloud-filtered (cloud radiance fraction <0.5) tropospheric NO<sub>2</sub> columns retrieved by *Martin et al.* [2006] for May 2003 to April 2005. The retrieval involves a spectral fit over 429–452 nm to produce total slant columns, removal of stratospheric NO<sub>2</sub> columns using SCIAMACHY observations over the Pacific, and an air mass factor calculation to correct for atmospheric scattering. The data have been validated with in situ airborne observations of NO<sub>2</sub> over and downwind of North America as part of the ICARTT campaign. The retrieval uncertainty is ±(5 × 10<sup>14</sup> molecules cm<sup>-2</sup> + 30%) for cloud-filtered scenes with larger uncertainty for cloudy scenes. The omission of cloudy scenes is of little consequence for our analysis due to the long upper tropospheric NO<sub>x</sub> lifetime of 5–10 days as inferred by *Jaeglé et al.* [1998].

[14] Figure 2 shows a seasonal average of cloud-filtered SCIAMACHY tropospheric NO<sub>2</sub> columns. Tropospheric NO<sub>2</sub> columns are strongly correlated with surface emissions [*Martin et al.*, 2003a; *Toenges-Schuller et al.*, 2006]. NO<sub>2</sub> columns are largest in winter over industrial regions due to seasonal variation in the NO<sub>x</sub> lifetime [*Velders et al.*, 2001]. The seasonal variation of tropical NO<sub>2</sub> columns reflects biomass burning and soil NO<sub>x</sub> emissions [*Jaeglé et al.*, 2005]. Identification of a lightning signal in satellite observations of NO<sub>2</sub> columns is complicated by the higher measurement sensitivity to NO<sub>x</sub> in the lower troposphere than in the upper troposphere [*Martin et al.*, 2002a], resulting from the factor of 25 increase in the NO/NO<sub>2</sub> ratio from the surface to the tropopause [*Bradshaw et al.*, 1999] that is driven by the temperature dependence of the NO + O<sub>3</sub> reaction.

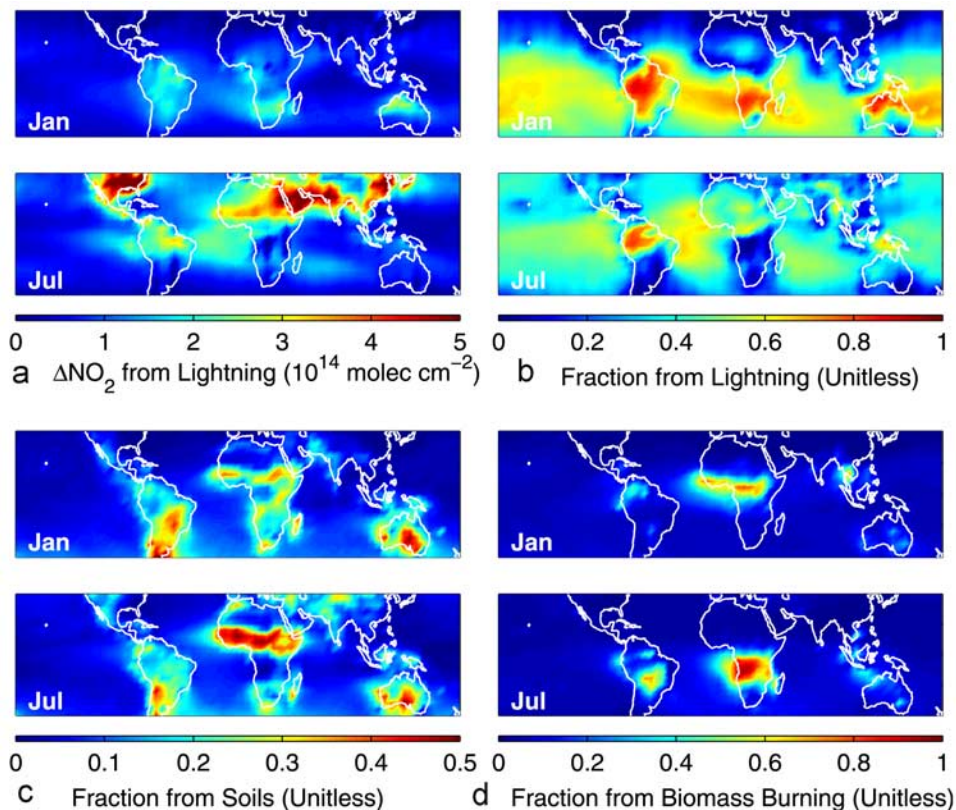




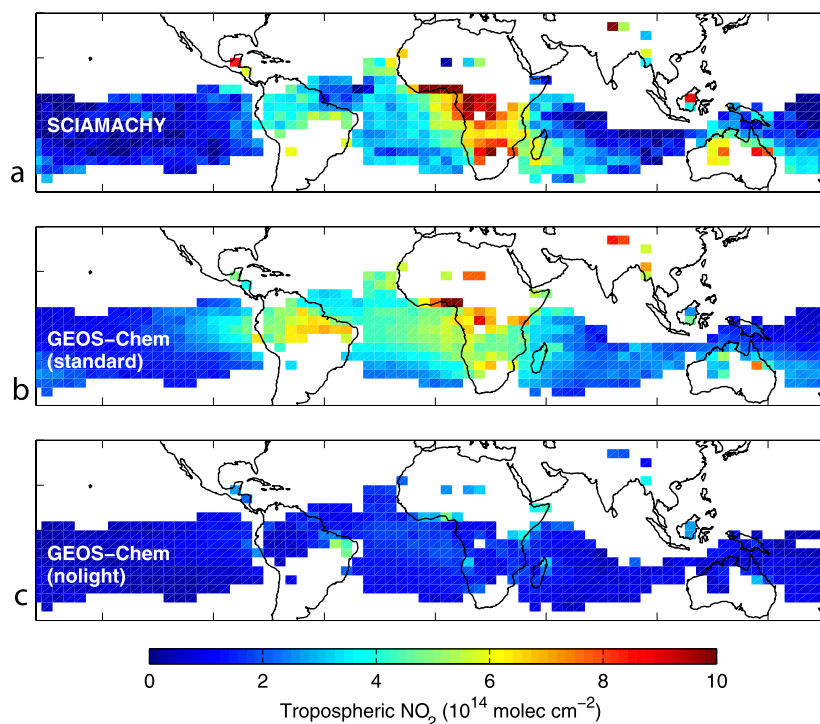
**Figure 2.** Mean tropospheric NO<sub>2</sub> columns retrieved from the SCIAMACHY satellite instrument for (top) November–April and (bottom) May–October. Scenes where clouds or snow dominate solar backscatter have been excluded from the average to reduce retrieval uncertainty. Data from *Martin et al.* [2006].

[15] The interpretation of satellite observations of tropospheric columns warrants clarification of the tropical tropopause. The level of zero radiative heating occurs at 14–15 km, above which air either mixes with the extratropical stratosphere or slowly ascends through the

thermal tropopause at 16–17 km and into the tropical stratosphere [*Highwood and Hoskins, 1998; Folkins et al., 1999*]. We refer to the tropopause as the thermal tropopause and to tropospheric columns as values below the thermal tropopause.



**Figure 3.** Sensitivity of simulated tropospheric NO<sub>2</sub> columns to NO<sub>x</sub> emissions from lightning, soils, and biomass burning for January and July. (a) Shown is the NO<sub>2</sub> column due to lightning NO<sub>x</sub> emissions as determined by the difference between the standard simulation and a simulation that excludes lightning NO<sub>x</sub> emissions. Also shown is the fraction of the simulated tropospheric NO<sub>2</sub> column from (b) lightning, (c) soils, and (d) biomass burning.



**Figure 4.** Annual average of tropospheric NO<sub>2</sub> columns at locations and times in which more than 60% of the simulated NO<sub>2</sub> column is from lightning, and less than 25% of the simulated NO<sub>2</sub> column is from any surface NO<sub>x</sub> source (soils, biomass burning, fossil fuels, or biofuel). White areas indicate regions where these thresholds were not met. Shown are (a) data retrieved from the SCIAMACHY satellite instrument, (b) the standard simulation with 6 Tg N yr<sup>-1</sup> from lightning, and (c) a simulation without lightning NO<sub>x</sub> emissions.

[16] Given the strong relationship of tropospheric NO<sub>2</sub> columns with surface emissions, we use the GEOS-Chem model for guidance in identifying locations where a lightning influence would be expected. Figure 3 shows the sensitivity of tropospheric NO<sub>2</sub> columns to NO<sub>x</sub> emissions from either lightning, soils, or biomass burning as determined from the difference between the standard simulation and sensitivity simulations that exclude either source. Figure 3a shows that the largest lightning-induced enhancements of the tropospheric NO<sub>2</sub> column are found over and downwind of active convective regions. The maximum monthly mean enhancements are comparable to the SCIAMACHY retrieval uncertainty.

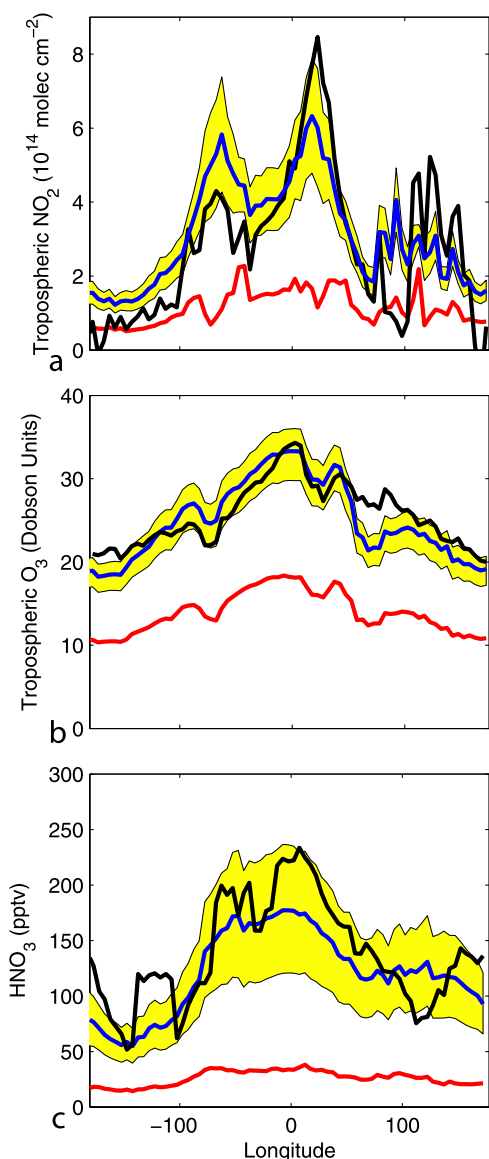
[17] Figure 3b shows the fraction of the NO<sub>2</sub> column from lightning. This fraction is anticorrelated ( $r = -0.49$ ) with the simulated tropical tropospheric NO<sub>2</sub> column, reflecting the greater sensitivity of SCIAMACHY to NO<sub>x</sub> in the lower troposphere than in the upper troposphere. The largest relative contributions from lightning are found over convective regions with negligible surface sources. Lightning has the broadest relative influence during January–April when flashes are most frequent in the Southern Hemisphere and when biomass burning is largely confined to the Northern Hemisphere. Lightning makes the weakest relative contribution during July–October when lightning is active in the Northern Hemisphere where there are numerous anthropogenic sources.

[18] Figures 3c and 3d show the fraction of the tropospheric NO<sub>2</sub> column from soil and biomass burning emis-

sions. The largest fractions from soils are found over tropical savannas during the wet season. Jaeglé *et al.* [2004] attributed GOME observations of tropospheric NO<sub>2</sub> column enhancements over northern Africa to rain-induced emissions of NO from soils. A large contribution from biomass burning of more than 50% is found over northern equatorial Africa during January and over central Africa during July. Soil and biomass burning emissions make a small contribution (typically <25%) to tropospheric NO<sub>2</sub> columns over oceans. The sum of the fractional contributions from these sources can exceed 100% due to nonlinearities.

[19] We sample the SCIAMACHY data at locations and months in which GEOS-Chem simulations indicate that more than 60% of the NO<sub>2</sub> column is from lightning, and less than 25% of the NO<sub>2</sub> column originates from any surface NO<sub>x</sub> source (soils, biomass burning, fossil fuel, or biofuel). These thresholds retain 15% of the tropical SCIAMACHY data. Most land regions are excluded by these thresholds, since surface NO<sub>x</sub> sources often dominate the NO<sub>2</sub> column over land.

[20] Figure 4a shows the annual average of the filtered SCIAMACHY tropospheric NO<sub>2</sub> columns. The largest enhancements are found over central Africa reflecting intense lightning activity [Christian *et al.*, 2003]. A broad enhancement of  $3\text{--}6 \times 10^{14}$  molecules cm<sup>-2</sup> is evident over the tropical Atlantic and Indian Oceans, reflecting upper tropospheric transport from continental regions. Lightning is the dominant source of reactive nitrogen transported into the tropical Atlantic in a numerical simulation by Moxim



**Figure 5.** Meridional average of annual mean trace gas concentrations for locations and months that are dominated by the effects lightning NO<sub>x</sub>. Shown are (a) tropospheric NO<sub>2</sub> columns for locations and months in which more than 60% of the tropospheric NO<sub>2</sub> column is from lightning, and less than 25% of the NO<sub>2</sub> column from any surface NO<sub>x</sub> source, (b) the tropospheric O<sub>3</sub> column averaged over locations and months in which more than 40% of the simulated concentrations are from lightning NO<sub>x</sub>, and (c) the HNO<sub>3</sub> mixing ratio between 200 and 350 hPa averaged over locations and months in which more than 60% of the simulated concentrations are from lightning NO<sub>x</sub>. Black lines indicate satellite observations. Blue lines indicate the standard simulation with 6 Tg N yr<sup>-1</sup> from lightning. The boundaries of the yellow shaded regions indicate sensitivity simulations of 4 and 8 Tg N yr<sup>-1</sup> from lightning. Red lines indicate the simulation without lightning NO<sub>x</sub> emissions.

and Levy [2000]. Previous analyses of tropospheric NO<sub>2</sub> columns retrieved from GOME also found evidence of lightning NO<sub>x</sub> over the tropical Atlantic [Richter and Burrows, 2002; Edwards et al., 2003; Boersma et al., 2005].

[21] Figure 4b shows the annual average of simulated tropospheric NO<sub>2</sub> columns, sampled at the same time and location as the SCIAMACHY measurements. The modeled NO<sub>2</sub> columns are broadly consistent with the SCIAMACHY data ( $r = 0.75$ ,  $n = 1937$ , bias = 8%). The simulation also exhibits a broad maximum over the tropical Atlantic Ocean and a broad minimum over the tropical Pacific Ocean. The simulation overestimates the lightning signal over South America and underestimates the lightning signal over Africa, common problems for models using the Price and Rind [1992] parameterization [Allen and Pickering, 2002]. This bias is reduced in the sensitivity simulation in which lightning flashes are rescaled to the OTD/LIS observations.

[22] Figure 4c shows the annual average of tropospheric NO<sub>2</sub> columns in the simulation without lightning NO<sub>x</sub> emissions. Tropospheric NO<sub>2</sub> columns in this simulation are biased low by 60% with respect to the SCIAMACHY data. Comparison with Figure 4b reveals that lightning largely explains the broad wave-1 pattern in the SCIAMACHY NO<sub>2</sub> columns.

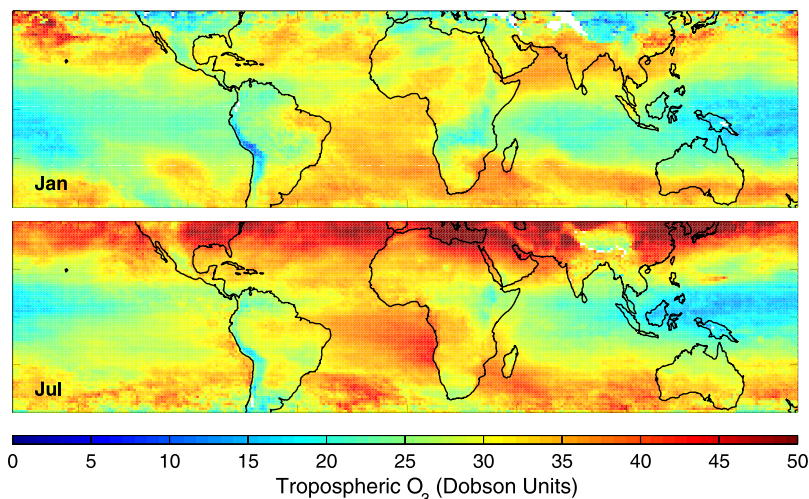
[23] Figure 5a compares the meridional average of all the tropospheric NO<sub>2</sub> columns in Figure 4. Both the SCIAMACHY NO<sub>2</sub> columns and the standard simulation show enhancements in the tropical Atlantic, relative to the tropical Pacific. No such pattern is found in the simulation without lightning NO<sub>x</sub> emissions. Emissions of 4–8 Tg N yr<sup>-1</sup> would best represent the SCIAMACHY data over most of the tropics. The magnitude of the annual mean lightning signal in the tropospheric NO<sub>2</sub> observations is about  $2\text{--}6 \times 10^{14}$  molecules cm<sup>-2</sup>, comparable to the local uncertainty of about  $5 \times 10^{14}$  molecules cm<sup>-2</sup> in the SCIAMACHY tropospheric NO<sub>2</sub> retrievals. However, it is likely that the retrieval error is reduced for the spatial and temporal mean examined here.

### 3.2. Analysis of Tropospheric Ozone Columns

[24] The OMI and MLS instruments were launched in July 2004 on board the Aura spacecraft into a polar Sun-synchronous orbit [Schoeberl et al., 2004]. OMI is a nadir-scanning instrument that detects backscattered solar radiance over 270–500 nm to measure column O<sub>3</sub> with near global coverage at a resolution of 13 km × 24 km at nadir [Levell et al., 2006]. The EOS MLS instrument is a thermal-emission microwave limb sounder that measures vertical profiles of mesospheric, stratospheric, and upper tropospheric O<sub>3</sub> from limb scans ahead of the Aura satellite [Froidevaux et al., 2006; Waters et al., 2006]. Ziemke et al. [2006] combined measurements from both instruments to retrieve daily tropospheric O<sub>3</sub> columns with a measurement uncertainty of 5 Dobson Units (DU) as determined by comparison with ozonesondes.

[25] Figure 6 shows tropospheric O<sub>3</sub> columns retrieved from the OMI and MLS instruments for January and July. A persistent wave-1 pattern is found in the southern tropics, with a maximum in the tropical Atlantic and a minimum in the tropical Pacific throughout the year [Fishman et al., 1990; Shiotani, 1992; Ziemke et al., 1996; Hudson and Thompson, 1998; Thompson et al., 2003; Sauvage et al., 2006]. The minimum in the tropical Pacific reflects the rapid destruction of O<sub>3</sub> in the marine boundary layer [Piotrowicz et al., 1991; Kley et al., 1996] coupled with





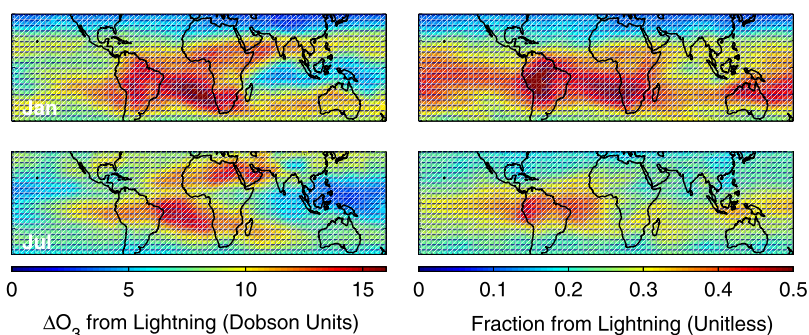
**Figure 6.** Tropospheric ozone columns retrieved from the Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder (MLS) for (top) January 2005 and (bottom) July 2005. Data from Ziemke *et al.* [2006].

the negative tendency of deep convection on the O<sub>3</sub> column [Lelieveld and Crutzen, 1994]. The persistent maximum over the tropical Atlantic reflects the integrated effect on O<sub>3</sub> of lightning emissions over the tropical continents [Jacob *et al.*, 1996; Martin *et al.*, 2002b; Sauvage *et al.*, 2007b], coupled with seasonal enhancements from biomass burning [Thompson *et al.*, 1996; Jenkins *et al.*, 1997]. The enhancement in the northern hemisphere during July is driven by photochemical production from anthropogenic emissions [Fishman and Brackett, 1997]. Enhancements at  $\pm 30^\circ$  are due to a combination of subsidence from the stratosphere and photochemical production [Li *et al.*, 2002; Jing *et al.*, 2005].

[26] Figure 7 shows the simulated sensitivity of tropospheric O<sub>3</sub> columns to lightning NO<sub>x</sub> emissions. Lightning makes a broad contribution to tropospheric O<sub>3</sub> columns throughout the tropics reflecting the upper tropospheric O<sub>3</sub> lifetime of several weeks. A particularly large O<sub>3</sub> column enhancement from lightning is apparent during January in the southern tropical Atlantic, contributing to the north-south oceanic gradient that was highlighted by Thompson

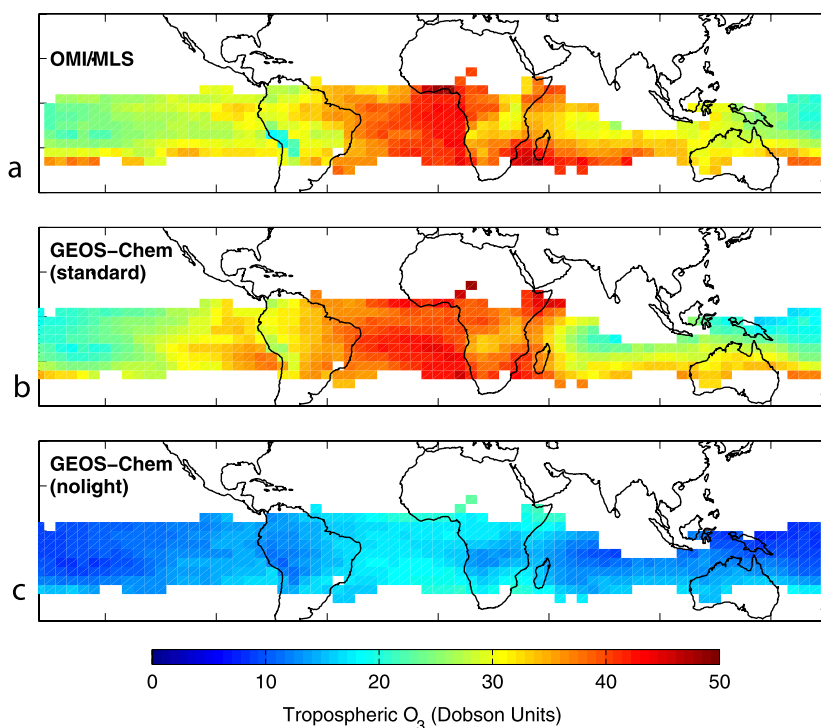
*et al.* [2000]. Weaker enhancements from lightning are found over the Middle East during July [Li *et al.*, 2001] and in the subsiding branches of the Hadley Circulation near  $\pm 30^\circ$ . Lightning makes a smaller relative contribution to the tropospheric O<sub>3</sub> column during July when biomass burning emissions are more abundant in the southern hemisphere and when lightning is predominantly in the northern hemisphere where anthropogenic sources dominate.

[27] Figure 8a shows the annual average over September 2004 to August 2005 of the OMI/MLS tropospheric O<sub>3</sub> columns at locations and times in which more than 40% of the simulated column is from lightning. This threshold retains 15% of the tropical OMI/MLS observations. Only 0.1% of the remaining observations would be affected by an additional threshold of 25% on the O<sub>3</sub> column fraction from surface sources. Figure 8b shows the annual average of the standard simulation sampled at the same locations and months as the OMI/MLS data. There is a high degree of consistency between the simulated and satellite data ( $r = 0.85$ ,  $n = 1953$ , bias =  $-1\%$ ). Figure 8c shows tropospheric



**Figure 7.** Sensitivity of tropospheric ozone columns to lightning NO<sub>x</sub> emissions for (top) January and (bottom) July. The left column indicates the tropospheric ozone column due to lightning NO<sub>x</sub> emissions as determined by the difference between the standard simulation and a simulation that excludes lightning NO<sub>x</sub> emissions. The right column indicates the corresponding fraction of the tropospheric ozone column from lightning NO<sub>x</sub> emissions.





**Figure 8.** Annual average of tropospheric ozone columns at locations and times in which GEOS-Chem simulations yield more than 40% of the tropospheric ozone column from lightning. White areas indicate regions where these thresholds were not met. Shown are (a) data determined from the OMI and MLS satellite instruments, (b) the standard simulation with  $6 \text{ Tg N yr}^{-1}$  from lightning, and (c) a simulation without lightning NO<sub>x</sub> emissions.

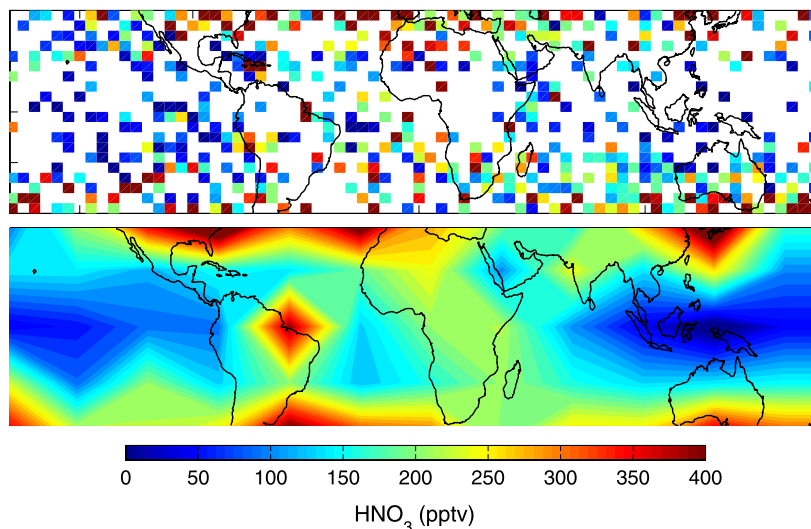
O<sub>3</sub> columns for the simulation without lightning NO<sub>x</sub> emissions, revealing a large negative bias of 45% versus the OMI/MLS observations. The magnitude of the lightning signal of 10–15 DU, as determined by the difference between the standard simulation and the simulation without lightning, is 2–3 times greater than the retrieval uncertainty.

[28] Figure 5b shows the meridional average of the tropospheric O<sub>3</sub> columns in Figure 8. Lightning NO<sub>x</sub> production

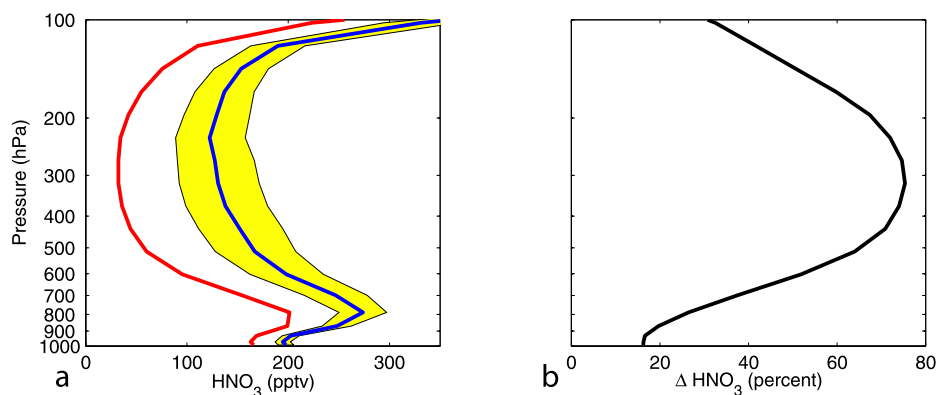
of 4–6 Tg N yr<sup>-1</sup> would best represent the OMI/MLS observations in the Atlantic, while a rate of 6–8 Tg N yr<sup>-1</sup> would best represent observations in the Pacific.

### 3.3. Analysis of Upper Tropospheric HNO<sub>3</sub>

[29] The ACE-FTS instrument [Bernath *et al.*, 2005] was launched on board the SCISAT-1 spacecraft into a low Earth orbit (altitude 650 km, inclination 74°) in August 2003.



**Figure 9.** Mean HNO<sub>3</sub> retrieved from the ACE-FTS satellite instrument over 200–350 hPa for March 2004 to February 2006 inclusive. The data are binned onto a 4° latitude by 5° longitude grid in the top panel and onto a 20° latitude by 30° longitude grid in the bottom panel. Data from Boone *et al.* [2005].



**Figure 10.** Vertically resolved sensitivity of the tropical (30°S–30°N) HNO<sub>3</sub> concentration to NO<sub>x</sub> emissions from lightning, showing (a) the annual mean tropical HNO<sub>3</sub> mixing ratio. The blue line indicates the standard simulation. The boundaries of the yellow shaded region indicate sensitivity simulations of 4 and 8 Tg N yr<sup>-1</sup> from lightning. The red line indicates simulated values in the simulation without lightning NO<sub>x</sub> emissions. Also shown is (b) the fraction of HNO<sub>3</sub> from lightning as determined by the difference between the standard simulation and a simulation without lightning NO<sub>x</sub> emissions.

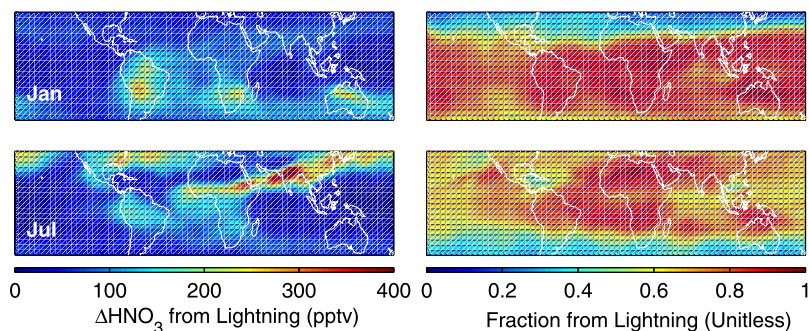
The ACE-FTS instrument is a high spectral resolution (0.02 cm<sup>-1</sup>) Fourier Transform Spectrometer operating over 750–4400 cm<sup>-1</sup> (2.2–13.3 μm). From the ACE-FTS solar occultation measurements, HNO<sub>3</sub> profiles are retrieved with an altitude resolution of 3–4 km [Boone *et al.*, 2005]. The retrievals employ HNO<sub>3</sub> spectral features in the ranges 860–910 cm<sup>-1</sup> and 1690–1730 cm<sup>-1</sup>. The typical measurement uncertainty in the upper troposphere is 35 pptv. The data used here are research retrievals for HNO<sub>3</sub>, a refinement of the version 2.2 ACE-FTS data set.

[30] Figure 9 shows the average of HNO<sub>3</sub> concentrations over 200–350 hPa from ACE-FTS for March 2004 to February 2006 inclusive. Enhancements are found poleward of 25° due to a combination of seasonally varying tropospheric and stratospheric sources. Consistently low values are seen over the Pacific Ocean. As with NO<sub>2</sub> and O<sub>3</sub>, we use the GEOS-Chem model to provide insight into the processes affecting the HNO<sub>3</sub> distribution.

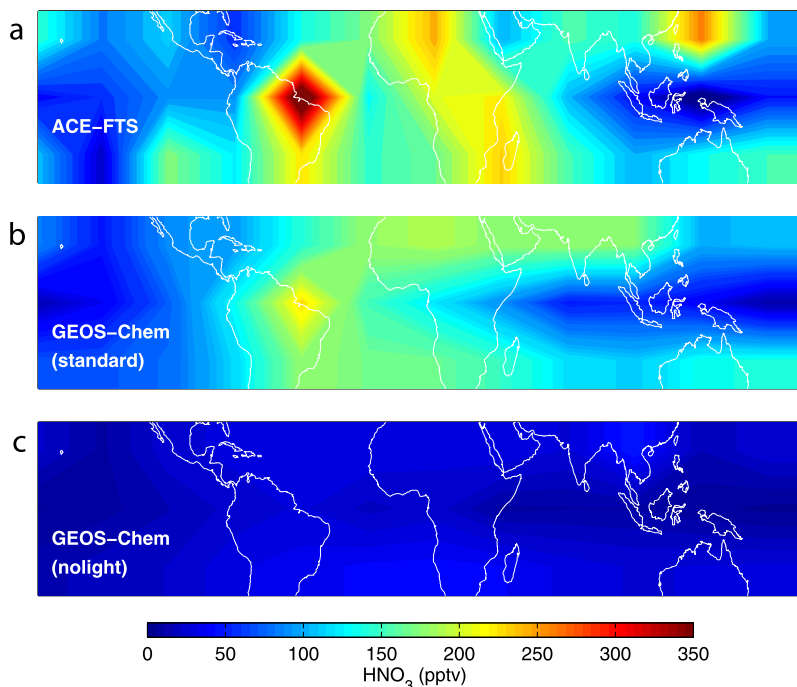
[31] We first examine the vertical sensitivity of HNO<sub>3</sub> to lightning NO<sub>x</sub> emissions. Figure 10a shows HNO<sub>3</sub> concentrations for simulations with and without lightning. A broad minimum in the tropical upper troposphere has been attributed to convective outflow of air depleted of this highly soluble

species [Folkins *et al.*, 2006]. The increase in HNO<sub>3</sub> with decreasing pressure represents an increasing stratospheric contribution, while the increase toward the surface is driven by photochemical production of HNO<sub>3</sub> during subsidence. The role of surface NO<sub>x</sub> sources increases markedly below 600 hPa. Figure 10b shows that oxidation of lightning NO<sub>x</sub> explains nearly 80% of HNO<sub>3</sub> concentrations over 200–350 hPa. We focus our attention on this region.

[32] Figure 11 shows the calculated horizontal sensitivity of HNO<sub>3</sub> concentrations to lightning NO<sub>x</sub>. The left column of Figure 11 indicates pronounced enhancements over and downwind of convective regions, especially in the southern tropics during January. The right column of Figure 11 indicates that during January lightning NO<sub>x</sub> explains 80% of HNO<sub>3</sub> mixing ratios over a broad region south of 25°N, with stratospheric NO<sub>y</sub> making a larger contribution further north. During July, stratospheric NO<sub>y</sub> plays a more important role at these pressures south of 25°S. Airborne measurements in the upper troposphere provide evidence that HNO<sub>3</sub> increases the relative humidity in low-temperature cirrus clouds [Gao *et al.*, 2004]. The pronounced contribution of lightning to upper tropospheric HNO<sub>3</sub> found here suggests that a hypothesized increase in lightning NO<sub>x</sub>



**Figure 11.** Sensitivity of HNO<sub>3</sub> concentrations over 200–350 hPa to lightning NO<sub>x</sub> emissions for (top) January and (bottom) July. The left column shows the HNO<sub>3</sub> mixing ratio due to lightning NO<sub>x</sub>, as determined by the difference between the standard simulation and a simulation without lightning NO<sub>x</sub> emissions. The right column shows the corresponding fraction of the HNO<sub>3</sub> mixing ratio from lightning.



**Figure 12.** Annual average of the HNO<sub>3</sub> mixing ratio at locations and times in which more than 60% of the simulated HNO<sub>3</sub> concentration is from lightning. Shown are (a) data retrieved from the ACE satellite instrument, (b) the standard simulation with 6 Tg N yr<sup>-1</sup> from lightning, and (c) a simulation without lightning NO<sub>x</sub> emissions.

emissions from climate change [Price and Rind, 1994] could have implications for upper tropospheric water vapor.

[33] Figure 12 shows the HNO<sub>3</sub> mixing ratio at locations and times in which more than 60% of the simulated HNO<sub>3</sub> is from lightning. This threshold retains 83% of the tropical ACE-FTS measurements while eliminating the seasonally varying stratospheric air poleward of 25°. The measured and simulated HNO<sub>3</sub> mixing ratios in Figures 12a and 12b are generally consistent ( $r = 0.75$ ,  $n = 360$ , bias = -12%). The simulation underestimates observed upper tropospheric HNO<sub>3</sub> over South America and Africa. However, both the simulation and the observations exhibit a maximum in the tropical Atlantic and a minimum in the tropical Pacific. This wave-1 pattern reflects the frequent injection of HNO<sub>3</sub> depleted air into the upper troposphere of the tropical Pacific, coupled with photochemical production during transport to the tropical Atlantic as part of the Walker circulation. Figure 12c shows that HNO<sub>3</sub> mixing ratios in the simulation without lightning NO<sub>x</sub> emissions are only 1/5th of the observations.

[34] Figure 5c shows the meridional average of the upper tropospheric HNO<sub>3</sub> in Figure 12. The simulation with 6 Tg N yr<sup>-1</sup> from lightning best represents the ACE-FTS observations over most of the world. The strength of the lightning signal in the observations ranges from a minimum of 50 pptv over the western Pacific Ocean to 200 pptv over the tropical Atlantic, exceeding the typical ACE-FTS measurement precision of 35 pptv.

#### 4. Discussion

[35] On balance, all three measurements yield a top-down estimate on the lightning NO<sub>x</sub> source of 4–8 Tg N yr<sup>-1</sup>,

with 6 Tg N yr<sup>-1</sup> being most likely. Potential errors in the simulations used to reach this conclusion are reduced by using three different trace gases with lifetimes ranging from several days for NO<sub>x</sub> to several weeks for O<sub>3</sub> and HNO<sub>3</sub>. Varying the thresholds used to filter the observations has little effect on the estimate. The sensitivity simulation in which the spatial distribution of lightning flashes is scaled to observations from the OTD and LIS instruments has negligible implications for the top-down estimate for all three trace gases, also supporting the conclusion. Annual mean tropospheric O<sub>3</sub> columns over the regions affected by lightning in the sensitivity simulation with enhanced intracloud lightning exhibit little difference (<1 DU) with the standard simulation, again supporting the conclusion. The small O<sub>3</sub> column enhancement in the simulation with enhanced intracloud lightning reflects compensating changes in upper and lower tropospheric O<sub>3</sub> and in its regional distribution. Savage *et al.* [2007a] further discuss the implications of both sensitivity simulations on tropospheric O<sub>3</sub>. The weak interannual variation in tropical tropospheric O<sub>3</sub> columns of less than 10% found by Ziemke and Chandra [1999] implies a similarly small interannual variation in the lightning NO<sub>x</sub> source.

[36] The global estimate of 6 ± 2 Tg N yr<sup>-1</sup> derived here is well within the range of recent estimates in Table 1. Our estimate is also consistent with the value of 5 ± 3 Tg N yr<sup>-1</sup> that is used in numerous chemical transport models as summarized by Zhang *et al.* [2003]. A global source strength of less than 4 Tg N yr<sup>-1</sup> or greater than 8 Tg N yr<sup>-1</sup> would imply gross errors in current simulations of tropospheric O<sub>3</sub> and reactive nitrogen.

[37] The sensitivity simulation with enhanced intracloud lightning NO<sub>x</sub> indicates that tropospheric NO<sub>2</sub> columns and

upper tropospheric HNO<sub>3</sub> concentrations could provide insight into the vertical profile of NO<sub>x</sub> emissions. Owing to the increasing NO/NO<sub>2</sub> ratio with altitude, tropospheric NO<sub>2</sub> columns in the simulation with enhanced intracloud lightning are about half of those in the standard simulation, well below the local retrieval uncertainty. In contrast, upper tropospheric HNO<sub>3</sub> concentrations in the simulation with enhanced intracloud lightning are 50–100% larger than in the standard simulation. These divergent effects could imply that in the tropics cloud-ground lightning releases more NO<sub>x</sub> than intracloud lightning. *Sauvage et al.* [2007a] similarly find that enhanced intracloud lightning NO<sub>x</sub> reduces the consistency of simulated O<sub>3</sub> versus measured vertical profiles from aircraft and ozonesondes. However, if enhanced tropical intracloud lightning is appropriate, two possible explanations for the bias in simulated HNO<sub>3</sub> are an underestimate in the GEOS-4 cloud mass flux detrainment profile [*Folkens et al.*, 2006], or the uptake of HNO<sub>3</sub> by ice in the upper troposphere [e.g., *Abbatt*, 1997; *Zondlo et al.*, 1997]. These coupled issues should be revisited with improved representations of vertical transport, when the existing uncertainty [*Ullerstam et al.*, 2005; *von Kuhlmann and Lawrence*, 2006] in the ice uptake process is reduced, and when the next generation of vertical profiles of lightning NO<sub>x</sub> production become available.

[38] The top-down estimate inferred here is based largely on tropical measurements. Included in this global estimate is a northern midlatitude lightning NO<sub>x</sub> source of 1.6 Tg N yr<sup>-1</sup> that is based on recent evidence from aircraft, ozonesonde, and satellite observations as part of the ICARTT and STERAO-A campaigns [*DeCaria et al.*, 2005; *Cooper et al.*, 2006; *Hudman et al.*, 2007; *Martin et al.*, 2006; *Pickering et al.*, 2006]. Recent estimates of the number of moles of NO released per flash over Germany are 70–80% of the North American values [*Fehr et al.*, 2004; *Ott et al.*, 2007] that were used for the midlatitude estimate. Future refinement of the midlatitude lightning NO<sub>x</sub> source will propagate into the global estimate.

[39] The OMI instrument also provides the capability for retrieval of tropospheric NO<sub>2</sub> columns [e.g. *Bucsela et al.*, 2006]. Unfortunately, the OMI tropospheric NO<sub>2</sub> product was not available to us at the time of this analysis. Future extension of this approach to use tropospheric NO<sub>2</sub> columns from OMI could benefit from its small footprint in the nadir and daily global coverage.

## 5. Conclusions

[40] We have analyzed space-based observations of tropospheric NO<sub>2</sub> columns from SCIAMACHY over 2003–2005, tropospheric O<sub>3</sub> columns from OMI and MLS over 2004–2005, and upper tropospheric HNO<sub>3</sub> from ACE-FTS over 2004–2006 to provide top-down constraints on lightning NO<sub>x</sub> emissions. A global chemical transport model (GEOS-Chem) was used to identify the locations and months in which lightning would be expected to make a significant contribution to each species. The satellite retrievals were sampled at those locations and months for comparison with model simulations using lightning NO<sub>x</sub> emissions of different magnitudes. This approach exploits the role of transport and photochemistry in separating the effect of lightning NO<sub>x</sub> from surface NO<sub>x</sub> sources. Partic-

ular attention was devoted to excluding regions of biomass burning and soil NO<sub>x</sub> emissions that could be misinterpreted as a lightning signal in the SCIAMACHY NO<sub>2</sub> columns.

[41] All three filtered satellite datasets revealed a prominent wave-1 pattern with a maximum in the tropical Atlantic and a minimum in the tropical Pacific. The wave-1 pattern is driven by injection of lightning NO into the upper troposphere over the tropical continents, followed by photochemical production of NO<sub>2</sub>, HNO<sub>3</sub>, and O<sub>3</sub> during transport to the tropical Atlantic as part of the Walker Circulation. The lightning signal over the tropical Atlantic and Africa was  $2\text{--}6 \times 10^{14}$  molecules NO<sub>2</sub> cm<sup>-2</sup>, 15 Dobson Units of O<sub>3</sub>, and 125 pptv HNO<sub>3</sub>, as determined by the difference between the standard simulation with 6 Tg N yr<sup>-1</sup> from lightning and a simulation without lightning NO<sub>x</sub> emissions. The lightning signal over the tropical Pacific was 25–75% weaker.

[42] Sensitivity simulations were conducted with the GEOS-Chem model for lightning NO<sub>x</sub> emissions of 0, 4, 6, and 8 Tg N yr<sup>-1</sup>. The simulations were sampled at the same locations and times as the satellite observations. Comparison of these simulations with the satellite observations reveals that it is very likely that global lightning NO<sub>x</sub> emissions are between 4 and 8 Tg N yr<sup>-1</sup>. Emissions of 6 Tg N yr<sup>-1</sup> provide the most consistent representation of the NO<sub>2</sub>, HNO<sub>3</sub>, and O<sub>3</sub> measurements. This conclusion is robust to scaling the spatial distribution of the simulated lightning NO<sub>x</sub> production to flash count observations from the OTD and LIS instruments.

[43] Tropospheric O<sub>3</sub> columns provide a particularly strong constraint on the magnitude of lightning NO<sub>x</sub> emissions due to their insensitivity to the relative vertical profile of lightning NO<sub>x</sub> production, and due to the large lightning signal of 10–15 DU that is 2–3 times greater than the measurement uncertainty. Tropospheric NO<sub>2</sub> columns and upper tropospheric HNO<sub>3</sub> concentrations provide additional information on the vertical profile of lightning NO<sub>x</sub> production. Increasing the altitude of lightning NO<sub>x</sub> production reduces the tropospheric NO<sub>2</sub> column by increasing the NO/NO<sub>2</sub> ratio, while increasing the upper tropospheric HNO<sub>3</sub> concentration by reducing convective scavenging. A sensitivity simulation with enhanced intracloud lightning yields NO<sub>2</sub> columns that are half of those in the standard simulation and HNO<sub>3</sub> concentrations that are 50–100% larger than those in the standard simulation.

[44] This analysis could be improved in a number of ways. The lightning signal is comparable to the measurement uncertainty for tropospheric NO<sub>2</sub>; a more accurate retrieval of this species would be particularly valuable. The ACE-FTS HNO<sub>3</sub> measurements remain sparse; additional measurements from the Tropospheric Emission Spectrometer (TES), the Microwave Limb Sounder (MLS), or the Interferometric Monitor for Greenhouse gases (IMG) instruments would be useful. Model development to better represent processes such as lightning, convection, pulsing of soil NO<sub>x</sub> emissions, and uptake of HNO<sub>3</sub> on ice would improve the robustness of the constraint. Development of an adjoint model would provide a more formal approach to extend this analysis.

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