The impact of North American anthropogenic emissions and lightning on long-range transport of trace gases and their export from the continent during summers 2002 and 2004

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[1] We analyze the contribution of North American (NA) lightning and anthropogenic emissions to ozone concentrations, radiative forcing, and export fluxes from North America during summers 2002 and 2004 using the University of Maryland Chemical Transport Model (UMD-CTM) driven by GEOS-4 reanalysis. Reduced power plant emissions (NO_x SIP Call) and cooler temperatures in 2004 compared to 2002 resulted in lower ambient ozone concentrations over the eastern United States. Lightning flash rates in early summer 2004 were 50% higher than 2002 over the United States. Over the North Atlantic, changes in ozone column between early summer 2002 and 2004 due to changes in lightning and meteorology exceeded the change due to emission reductions by a factor of 7. Late summer changes in lightning had a much smaller impact on ozone columns. In summer 2004, net downward radiative flux at the tropopause due to ozone produced from anthropogenic emissions ranged from 0.15 to 0.30 W m^{-2} across the North Atlantic, while that due to ozone produced from lightning NO emissions ranged from 0.20 to 0.50 W m^{-2}. Enhanced lofting of polluted air followed by stronger westerly winds led to more net export of NO_x, NO_y, and ozone in early summer 2004 than 2002 despite reduced anthropogenic emissions. Ozone export fluxes across the eastern NA boundary due to anthropogenic emissions were factors of 1.6 and 2 larger than those due to lightning in 2004 and 2002, respectively. Doubling the NA lightning NO source increased downwind ozone enhancements due to lightning NO emissions by one third.

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1. Introduction

[2] North America is a major source of anthropogenic and naturally generated trace gases, and North American (NA) emissions affect trace gas mixing ratios over the North Atlantic, Europe and North Africa [*Li et al.*, 2002]. A key trace gas for both chemistry and radiative balance of the troposphere is ozone (O_3). According to the *Intergovernmental Panel on Climate Change* (*IPCC*) [2007], tropospheric O_3 is the third most important anthropogenic climate gas. Major precursors of tropospheric O_3 are nitrogen oxides (NO_x = NO + NO_2) from fuel combustion, soils and lightning. Surface precursors are rapidly transported upward via convection [*Dickerson et al.*, 1987; *Pickering et al.*, 1992, 1995] and detrained into the upper troposphere (UT) [*Bertram et al.*, 2007] where concurrent lightning greatly enhances NO_x [*DeCaria et al.*, 2000, 2005; *Zhang et al.*, 2003; *Hudman et al.*, 2007]. The importance of the vertical distribution of O_3 and its precursors is emphasized by the fact that mid-tropospheric and upper tropospheric O_3 has larger radiative forcing efficiency [*Lacis et al.*, 1990] than O_3 in the lower troposphere (LT). Therefore, O_3 resulting from lightning NO_x and NO_x transported upward in deep convection has the greatest consequences for the greenhouse effect [*IPCC*, 2007].

[3] The longer chemical lifetimes and greater wind speeds aloft can then lead to significant long-range transport (LRT) during which photochemical O_3 production occurs. On the other hand, the vertical mixing that occurs during convection over unpolluted regions can decrease the tropospheric O_3 column as high-O_3 air from the UT is transported down-

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ward to levels where it is destroyed more quickly, and low-O_3 air that originated near the surface is deposited in the UT [*Lelieveld and Crutzen*, 1994].

[4] The measurements between 1 July and 15 August 2004 from the INTEX-A (Intercontinental Chemical Transport Experiment–Phase A) aircraft campaign over the contiguous United States and adjacent areas [*Singh et al.*, 2006] and from coordinated IONS (INTEX Ozonesonde Network Study) ozonesondes launches [*Thompson et al.*, 2007a, 2007b] showed that the NA UT was greatly influenced by both NO_x from lightning (LNO_x) and surface pollution lofted via convection and contained elevated concentrations of peroxyacetylnitrate (PAN), O_3, hydrocarbons and NO_x [*Singh et al.*, 2007; *Cooper et al.*, 2006].

[5] *Hudman et al.* [2009] found that during the INTEX-A period the hemispheric tropospheric O_3 burden was enhanced with comparable contributions from anthropogenic and lightning NO emissions over North America. *Choi et al.* [2009] reported that LNO_x has a greater impact on radiation via O_3 production than its anthropogenic counterpart (ANO_x) over North America. Modeling of the horizontal and vertical distribution of LNO_x is highly uncertain. In the study by *Hudman et al.* [2007], the GEOS-Chem standard simulation greatly underestimated NO_x in the UT. After increasing the lightning NO production to 500 mol flash^{-1}, GEOS-Chem simulated NO_x was still low biased. Similarly, *Bousserez et al.* [2007], *Pierce et al.* [2007], *Fang et al.* [2010], and *Allen et al.* [2010] underestimated upper tropospheric NO_x using the MOCAGE, RAQMS, MOZART and GMI chemical transport models (CTMs) (all with different lightning schemes), respectively.

[6] We extend the previous work focused on NO_y export from North America during the INTEX-A period in summer 2004 by estimating the climate implications (radiative effects), by contrasting the summer 2004 with a meteorologically different summer (2002) using the University of Maryland Chemical Transport Model (UMD-CTM) [*Park et al.*, 2004a, 2004b]. *Godowitch et al.* [2008], using the Community Multiscale Air-Quality (CMAQ) model, showed that reduced NO_x emissions from power plants [*Frost et al.*, 2006; *Kim et al.*, 2006] caused substantial decreases in NO_x concentrations aloft (300–1100 m) and in ground level daily 8 h maximum O_3 between the summers 2002 and 2004. Sites downwind of the emission-rich Ohio River Valley (ORV) region (Pennsylvania, Ohio, West Virginia, Kentucky, Indiana and Illinois) experienced the greatest decreases in daily maxima of 8 h O_3 between 2002 and 2004. Interestingly, *Godowitch et al.* [2008] found that meteorological effects had greater impact on O_3 than those from emission changes over the region north of the Ohio River (Illinois, Indiana, Ohio, Wisconsin and Michigan). In particular, temperature and moisture parameters were considerably different in summer 2004 than 2002. Average maximum temperatures were substantially cooler in the northeastern United States, by as much as 3°C–5°C, during summer 2004 [*Godowitch et al.*, 2008]. Meteorology over northeastern North America during summer 2004 was dominated by a persistent low pressure, and there were increased synoptic disturbances relative to summer 2002 [*Thompson et al.*, 2007a, 2007b; *Büker et al.*, 2008]. The number of cold frontal passages over the northeastern United States was above average in summer 2004 [*Fuelberg et al.*, 2007]. As we estimate later the change in LNO_x emissions (due to more frequent lightning in summer 2004 than in 2002) is at least a factor of 2 larger than the change in ANO_x emissions (due to power plant NO_x reductions).

[7] In addition, we analyze the impact of the North American Monsoon. The monsoon region of the southwestern United States and northwestern Mexico does not have large ANO_x emissions but has a large increase in LNO_x emissions after the onset of the monsoon [*Ridley et al.*, 1994]. Much of the LNO_x becomes trapped in the UT above the Gulf of Mexico, the southern United States and Mexico—the major NA lightning region [*Li et al.*, 2005; *Cooper et al.*, 2006]—where conditions are favorable for O_3 production. On the basis of rainfall statistics over the southwestern United States (Arizona and New Mexico) and northwestern Mexico, 2004 is considered a weak monsoon year and 2002 is a near-normal or slightly weak monsoon year (daily climatology available at ftp://ftp.cpc.ncep.noaa.gov/precip/CPC_UNI_PRCP/GAUGE_GLB/).

[8] For both summers, we quantify the NA contribution to tropospheric O_3 by conducting sensitivity simulations with either anthropogenic or lightning emissions over North America shut off. In section 2, we describe the updated UMD-CTM, which has undergone major revision since *Park et al.* [2004a] and lightning simulations performed for this study. Section 3 includes model comparisons with aircraft, ozonesonde, satellite and ground-based measurements. We determine the model biases for O_3, NO_x and other trace gases. We then discuss the summer-to-summer variability of lightning and the radiative impact of O_3 produced from NA anthropogenic and lightning emissions in the outflow region. The results are summarized in section 4.

2. Model Description

[9] The UMD-CTM was described in detail by *Park et al.* [2004a]; here we describe it briefly in terms of the experimental design. The horizontal resolution of the model is 2° × 2.5°. From the surface to 9.3 hPa, there are 14 sigma layers and 17 constant pressure layers with a sigma pressure transition (at 177 hPa) near the tropopause. The UMD-CTM is driven by assimilated meteorological fields from version 4 of the Goddard Earth Observing System (GEOS-4) of the NASA Global Modeling and Assimilation Office. Specifically, we use the GEOS-4 CERES (Clouds and the Earth's Radiant Energy System) reanalysis (http://gmao.gsfc.nasa.gov/research/merra/sci_archive/climate.php). Convection in GEOS-4 [*Bloom et al.*, 2005] is represented by two parameterizations: deep convection follows *Zhang and McFarlane* [1995], while shallow convection is based on work by *Hack* [1994]. Moist convective transport in the UMD-CTM is parameterized using updraft, downdraft, entrainment and detrainment fields from the GEOS-4 CERES reanalysis. Turbulent mixing is calculated through a fractional mixing scheme [*Allen et al.*, 1996]: during a CTM time step (15 min) 20% of the mass in each model layer within the BL is mixed completely throughout the BL. Stratospheric O_3 flux into the troposphere is controlled through the synthetic O_3 (Synoz) scheme [*McLinden et al.*, 2000] as in work by *Park et al.* [2004a]. The Synoz-based flux is set to 475 Tg O_3 yr^{-1} for both years following *McLinden et al.* [2000].

Table 1. The UMD-CTM Simulations With Different Sources of NO_x Emissions

Simulation Name	Anthropogenic NO_x[a]	Lightning NO_x[b]	Period Simulated
L0	CEMS 2004	OTD/LIS (240)	May–Aug 2004
L1 (standard)	CEMS 2002, CEMS 2004	NLDN-based (240)	May–Aug 2002, May–Aug 2004
L2 (doubled lightning)	CEMS 2004	NLDN-based (480)	May–Aug 2004
noAnthro-NA[c]	none	NLDN-based (240)	May–Aug 2002, May–Aug 2004
noL-NA[c]	CEMS 2002, CEMS 2004	none	May–Aug 2002, May–Aug 2004
no NO_x SIP Call	CEMS 2002	NLDN-based (240)	May–Aug 2004

[a]Emission inventory used for the power plant sector for the contiguous United States (CONUS).
[b]Observed flash rates used to adjust the model-calculated flash rates over the CONUS; lightning NO moles produced per flash over the CONUS are in parentheses.
[c]North America is defined as Canada, the CONUS, Mexico, and the Gulf of Mexico.

[10] We use the same chemical mechanism as in the work by *Park et al.* [2004a] but with updated rate constants based on work by the *Jet Propulsion Laboratory* [2006]. We implemented the parameterization of quantum yields to update the photolysis rates for acetone on the basis of work by *Blitz et al.* [2004]. The wet deposition scheme [*Liu et al.*, 2001] includes contributions from scavenging in convective updrafts and rainout and washout from convective anvils and large-scale precipitation, and it allows for reevaporation.

[11] Table 1 identifies the modeling scenarios used to isolate the impacts of anthropogenic emissions, lightning and their summer-to-summer variability on O_3 concentrations. Initial conditions for O_3 were obtained from NASA's Global Modeling Initiative Chemical Transport Model (GMI CTM) [*Douglass et al.*, 2004] driven by meteorological input from the Finite Volume General Circulation Model (FVGCM) with several-year spin-up. Initial conditions for other species were obtained from a reduced 4° × 5° simulation of 1985 with the UMD-CTM by *Park et al.* [2004a] in the troposphere and from the GMI CTM in the stratosphere. The meteorological fields from the FVGCM do not correspond to a particular year.

2.1. Anthropogenic Emissions

[12] In 1998, the U.S. Environmental Protection Agency (EPA) issued a regulation to reduce the interstate transport of NO_x and ground level O_3 in the eastern United States [*Environmental Protection Agency*, 2005]. This rule, commonly known as the NO_x State Implementation Plan (SIP) Call, became effective in 2003 and required substantial power plant NO_x emission reductions in 22 eastern states [*Frost et al.*, 2006] with full implementation of controls to be completed by the summer 2004 O_3 season. In 2000, according to EPA's National Emission Inventory (NEI) and the Emission Database for Global Atmospheric Research (EDGAR), U.S. power generation accounted for one quarter (1.5 Tg N) of national ANO_x emissions (5.9 Tg N). Other major sources included road transport (1.9 Tg N), international shipping (0.6 Tg N) and air transport (0.3 Tg N).

[13] Global anthropogenic emissions in the model are as described by *Park et al.* [2004a] unless otherwise specified. Monthly power plant NO_x emissions from the United States are taken from Continuous Emission Monitoring System (CEMS). These direct measurements represent one of the most accurate parts of the U.S. emission database (http://www.epa.gov/airmarkets/emissions). All other anthropogenic emissions are from EDGAR 3.2 Fast Track 2000 (J. A. van Aardenne et al., The Edgar 3.2 Fast Track 2000 dataset (32FT2000), 2000, available at http://themasites.pbl.nl/images/Description_of_EDGAR_32FT2000(v8)_tcm61-46462.pdf) [*Olivier and Berdowski*, 2001; *Olivier et al.*, 2005]. Because of EPA's SIP Call, NO_x emitted from ORV power plants decreased on average by 50% between the summers 2002 and 2004. Overall, the NO_x SIP Call resulted in a 10% reduction in total ANO_x emissions from the contiguous United States (CONUS).

[14] The power plant NO_x emissions are released from tall stacks (average stack height is 76 m) in plumes with considerable buoyancy (average release temperature is 117°C). Stack emissions of NO_x are injected into the second-lowest model layer. All other anthropogenic emissions are injected into the lowest model layer. In the UMD-CTM, the lowest levels are centered at approximately 50, 250, 600, 1100 and 1900 m above the local surface.

[15] We increase ANO_x emissions in eastern China by 15% above the 2000 EDGAR NO_x emissions for both summers since a large positive trend of tropospheric NO_2 was reported by *Richter et al.* [2005] and *van der A et al.* [2006] over the industrial areas in China. It should be noted that we hold all nonpower plant U.S. NO_x emissions constant between 2002 and 2004; we also hold non-U.S. ANO_x emissions of any type constant between the 2 years. The spatial distribution of the changes in surface NO_x emissions from summer 2002 to 2004 over the United States used in the UMD-CTM simulations is shown in the auxiliary material (Figure S1).[1]

2.2. Lightning

[16] The annual LNO_x production is set to 5 Tg N yr^{-1}, which is in the center of the currently accepted range of 2–8 Tg N yr^{-1} [*Schumann and Huntrieser*, 2007]. The lightning scheme follows *Allen et al.* [2010]. The LNO_x production is assumed to be directly proportional to lightning flash rate *FR* as

$$FR = G \times L \times (zmmu - zzmu_0)^{\gamma},$$

where *zmmu* is GEOS-4 CERES upward cloud mass flux (at ~430 hPa). The lightning is thus colocated with the convective transport in the CTM. *FR* is set to zero for $zmmu < zzmu_0$. We use $zzmu_0 = 0.57$ kg m^{-2} min^{-1} as in the work by *Allen et al.* [2010] with $\gamma = 2$, thus assuming that the *FR* is a quadratic function of *zmmu*; $\gamma = 2$ gives more

[1]Auxiliary materials are available in the HTML. doi:10.1029/2010JD014305.

Table 2. The Lightning Sources Used in This Study and Other Studies

Reference	Scaling[a]	Lightning Source[b]	Time Period
This study, simulation L0	OTD/LIS	0.16 (240)	1 Jul to 15 Aug 2004
This study, simulation L1	NLDN	0.25 (240)	1 Jul to 15 Aug 2004
This study, simulation L2	NLDN	0.50 (480)	1 Jul to 15 Aug 2004
Hudman et al. [2007, 2009]	no scaling	0.27 (500)	1 Jul to 15 Aug 2004
Jourdain et al. [2010], simulation L0 (base)[c]	OTD/LIS	0.15 (260)	1 Jul to 15 Aug 2006
Jourdain et al. [2010], simulation L1 (NLDN)[c]	NLDN	0.21 (260)	1 Jul to 15 Aug 2006
Jourdain et al. [2010], simulation L2 (ligh×2)[c]	NLDN	0.42 (520)	1 Jul to 15 Aug 2006
Allen et al. [2010], simulation L0 (low NO_x)	OTD/LIS	0.17 (240)	1 Jul to 15 Aug 2004
Allen et al. [2010], simulation L0 (high NO_x)	OTD/LIS	0.34 (480)	1 Jul to 15 Aug 2004

[a]Observed flash rates used to adjust the model-calculated (different models and lightning schemes) flash rates.
[b]The lightning NO_x source in Tg N from the contiguous United States and adjacent coastal areas; lightning NO moles produced per flash are in parentheses.
[c]Please note the different year (2006). The lightning sources for the period of 1 July to 15 August are estimated by multiplying the lightning source values from July 2006 by a factor of 1.5.

realistic day-to-day variability in model-calculated flash rates, decreases biases and improves correlations with respect to observed flash rates than $\gamma = 1$. Using observations from the spaceborne Optical Transient Detector/Lightning Imaging Sensor (OTD/LIS) [*Boccippio et al.*, 2002; *Christian et al.*, 2003; *Mach et al.*, 2007] and from the ground-based National Lightning Detection Network (NLDN) [*Cummins et al.*, 1998; *Orville and Huffines*, 2001], we scale lightning flash rates (*FR*) globally (*G*) and locally (*L*) so the model flash rates per grid box match the NLDN and/or OTD/LIS observed data sets on a monthly basis (details in the work by *Allen et al.* [2010], who scaled *FR* to match the global OTD/LIS v2.2 climatology). In the vertical, we partition lightning NO emissions on the basis of the modeling studies of *Pickering et al.* [1998].

[17] Table 1 shows three lightning simulations with the UMD-CTM.

[18] 1. In L0, total flash rates (*FR*) derived from convective mass fluxes are adjusted to match the flash rates observed by OTD/LIS from space. We use Low Resolution Monthly Time Series (LRMTS) in the region between 35°S and 35°N and Low Resolution Annual Climatology (LRAC) elsewhere (available at http://thunder.msfc.nasa.gov/data). Since month-specific LIS observations are available only south of 35°N, simulation L0 does not account for summer-to-summer variability of NA lightning poleward of 35°N.

[19] 2. In L1, in addition to the L0 approach, over the CONUS, the flash rates derived from convective mass fluxes are adjusted to match the monthly average NLDN-based IC (intracloud) + CG (cloud-to-ground) flash rates (details are below). L1 is called "standard simulation."

[20] 3. In L2, in addition to the L1 approach, NO production per flash over the NA midlatitudes (25°N–50°N) is increased by a factor of 2 to 480 mol flash^{-1}, which nearly matches the estimates of *Ott et al.* [2010] derived from cloud-resolved modeling and of *Hudman et al.* [2007] used in their improved GEOS-Chem simulation of the INTEX-A period.

[21] When determining the NLDN-based IC + CG flash rates (simulations L1 and L2), we remove NLDN flashes with peak currents between 0 and 20 kA, since they are assumed to be IC in character [*Biagi et al.*, 2007]. We only use data over the CONUS for scaling as the NLDN detection efficiency drops off rapidly beyond 300 km from shore. The NLDN underwent a system-wide upgrade during 2002 [*Cummins et al.*, 2006]. The mean preupgrade detection efficiency over the CONUS was ~85%. After this upgrade, which began in spring 2002, the NLDN had a detection efficiency of 90–95% over the CONUS. For summer 2004, we thus use a detection efficiency of 93%. In order to estimate the detection efficiency for summer 2002, we average the preupgrade value derived from *Cummins et al.* [1998, Figure 9] and postupgrade value of 93%. To obtain the total IC + CG flash rates, we multiply the detection efficiency–adjusted NLDN CG flashes by $Z + 1$, where Z is the IC/CG ratio. *Boccippio et al.* [2001] constructed a 0.5° × 0.5° daily climatology of Z ratios (not year specific), by using observations of NLDN CG flashes and OTD/LIS total (IC + CG) flashes. In our study, we smooth their Z composite with a 7.5° moving boxcar, calculate the monthly averages and interpolate onto the 2° × 2.5° UMD-CTM grid. Before smoothing, we exclude grid boxes with $Z > 12$ as these values are anomalous [*Boccippio et al.*, 2001].

[22] To compare the lightning sources in our simulations with other investigators, we summarize the lightning NO emissions over the CONUS and adjacent coastal areas during INTEX-A (1 July to 15 August 2004) in Table 2. Simulations L0, L1 and L2 yield LNO_x emissions of 0.16 Tg N, 0.25 Tg N and 0.50 Tg N, respectively. *Hudman et al.* [2007, 2009], using a cloud top height–based flash rate scheme and assuming 500 NO mol flash^{-1}, obtained a LNO_x emission of 0.27 Tg N over the same areas for that period. They noted that their flash rates were biased low with respect to NLDN-based flash rates (assuming an IC/CG ratio of 3). Adjusting for this bias, they obtained a best estimate of 0.45 Tg N for the lightning NO source; however, they did not use this in their model simulations. *Jourdain et al.* [2010], with their GEOS-Chem simulation with NLDN-based flashes and an assumed production of 520 NO mol flash^{-1}, obtained a source of 0.28 Tg N for July 2006. Extrapolating to 1.5 months gives 0.42 Tg N, which is close to the bias-adjusted estimate by *Hudman et al.* [2007] for 2004. *Allen et al.* [2010], using the GMI CTM, reported 0.17 Tg N in their standard simulation and 0.34 Tg N in their simulation with doubled lightning NO production (480 mol flash^{-1}). They scaled to OTD/LIS climatology rather than NLDN-based flash rates. The magnitude of lightning source in our standard simulation (L1) nearly matches the one used by Hudman et al., while the L2 source (0.50 Tg N) is close to their NLDN-based estimate of the source.

[23] It is noteworthy that the 50% increase in CONUS LNO_x emissions between 2002 and 2004 more than offsets the ANO_x emission decreases due to the NO_x SIP Call. By applying this 50% change to L1 and L2 sources above, we obtain estimates of 0.13 and 0.25 Tg N, respectively, for the LNO_x emission changes from the same areas and time period as above. These estimated LNO_x emission changes are at least a factor of 2 larger than the corresponding change of 0.06 Tg N in ANO_x emissions due to the NO_x SIP Call (the ANO_x emissions from the CONUS were reduced from 0.57 Tg N to 0.51 Tg N) during the same time period. Of course the impact of the ANO_x emissions changes is most noticed near the surface while the impact of the LNO_x emissions changes is most important in the UT.

2.3. Biogenic Emissions

[24] Isoprene emissions used in the UMD-CTM simulations come from monthly average hourly emissions calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) from standard case for summer 2003 [*Guenther et al.*, 2006]. One of the most important meteorological factors in determining the isoprene emissions is the temperature. *Pacifico et al.* [2009, Figure 4] show that a 1°C temperature change can increase isoprene emissions by 15% for standard conditions (25°C–35°C). Temperatures in the region of high isoprene emissions were similar during summers 2002, 2003 and 2004 (see Figure S2 in the auxiliary material). Outside this region, maximum temperatures in 2002 exceeded maximum temperatures in 2004 by 1°C–5°C likely leading to more emissions in 2002 than in 2004. *Hogrefe et al.* [2004], in an isoprene sensitivity simulation with CMAQ, showed that summertime 8 h O_3 changed by <3 ppbv at locations within the domain (the eastern and central part of the United States) when isoprene emissions were increased by 20%–50% corresponding to maximum temperature increases of 1.5°C–3.5°C. *Nolte et al.* [2008], in another isoprene sensitivity simulation with CMAQ, showed that summertime 8 h O_3 increased by 1 ppbv or less over most of the CONUS, when isoprene emissions were increased by 25%. Therefore, the use of the same isoprene emissions for 2002 and 2004 is likely to have only a minor impact on conclusions from this study.

2.4. Biomass Burning

[25] Biomass burning emissions south of 48°N were derived from the Global Fire Emissions Database Version 2 (GFEDv2) [*van der Werf et al.*, 2006]. This data set prescribes emissions of total carbon as well as CO, CH_4 and NO_x. For other species, the total carbon emissions are converted to dry matter burned assuming a biomass carbon fraction of 0.45. Emission factors from *Andreae and Merlet* [2001] are then applied to estimate nonmethane hydrocarbon emissions. Factors are provided separately for savannah/grassland, tropical forest, extratropical forest and agricultural burning. Poleward of 48°N, we use emissions derived from Boreal Wildfire Emissions Model (BWEM) [*Kasischke et al.*, 2005] for summer 2002 and GFEDv2 emissions for summer 2004 (BWEM emissions for 2004 are unavailable). Land cover classification is derived from MODIS data [*Hansen et al.*, 2000; *Friedl et al.*, 2002].

[26] The GFEDv2 database uses the CASA model to estimate fuel loads [*van der Werf et al.*, 2003] and a burned area database derived from MODIS observations [*Giglio et al.*, 2006] to estimate monthly biomass burning emissions on a 1° × 1° grid. For this study, MODIS active fire data [*Justice et al.*, 2002] are used to calculate a daily perturbation for each 1° × 1° grid cell. This perturbation function is then applied to GFEDv2 emissions to obtain daily estimated emissions without altering monthly emissions, similar to the approach used by *Heald et al.* [2003]. This approach has been demonstrated to improve the accuracy of atmospheric simulations as opposed to using monthly averaged emissions [*Hyer et al.*, 2007; *Roy et al.*, 2007]. Biomass burning data at 1° × 1° resolution are smoothed with a 7 day moving average to reduce the effect of a periodic bias associated with the polar orbit of MODIS [*Heald et al.*, 2003] and are regridded onto the 2° × 2.5° UMD-CTM grid. Biomass burning emissions are injected below 1.5 km outside the tropics and below 0.5 km within the tropics (in vertical, both uniformly distributed by mass).

2.5. Radiative Forcing Calculation

[27] *IPCC* [2007] defines radiative forcing for tropospheric O_3 as the net downward flux (both the longwave and the much smaller shortwave contribution) at the tropopause due to the anthropogenic increase in tropospheric O_3 from preindustrial times; the global annual average present-day radiative forcing (stratospheric adjusted) due to tropospheric O_3 is +0.35 [−0.1, +0.3] W m^{-2} as estimated by climate simulations. If the stratospheric temperatures are not adjusted, then the forcing is called the instantaneous radiative forcing. While the IPCC definition only considers anthropogenic changes, in general, both anthropogenic and natural O_3 contribute to instantaneous radiative forcing (i.e., reduction in the outgoing longwave radiation). In our study, we consider the instantaneous radiative forcing of O_3 produced from anthropogenic emissions and lightning NO emissions separately and compare their relative effects during long-range transport of trace gases from North America.

[28] We calculate the longwave (980–1100 cm^{-1} band) contribution of the net downward Radiative Flux at the tropopause for clear-sky conditions (for brevity we refer to this as RF) from O_3 enhanced by anthropogenic emissions and lightning. RF serves as a measure of the extra heat (in W m^{-2}) input into the troposphere due to changes in O_3 (before stratospheric temperatures are adjusted to the radiative perturbation). We use the radiative transfer model from *Chou and Suarez* [1994], *Chou et al.* [1995] and *Park et al.* [2001]. Thermal infrared radiatively active constituents include N_2O, CH_4, CFC11, CFC12, CFC22, H_2O, CO_2, O_3 and background aerosol. Vertical distributions of O_3 are calculated by the UMD-CTM. Distributions of the other optically active constituents are held constant with respect to time. Water vapor and temperature profiles and skin temperatures are prescribed from the GEOS-4 CERES reanalysis.

3. Results

[29] In North America, O_3 concentrations and outflow are affected by both emission reductions and changes in

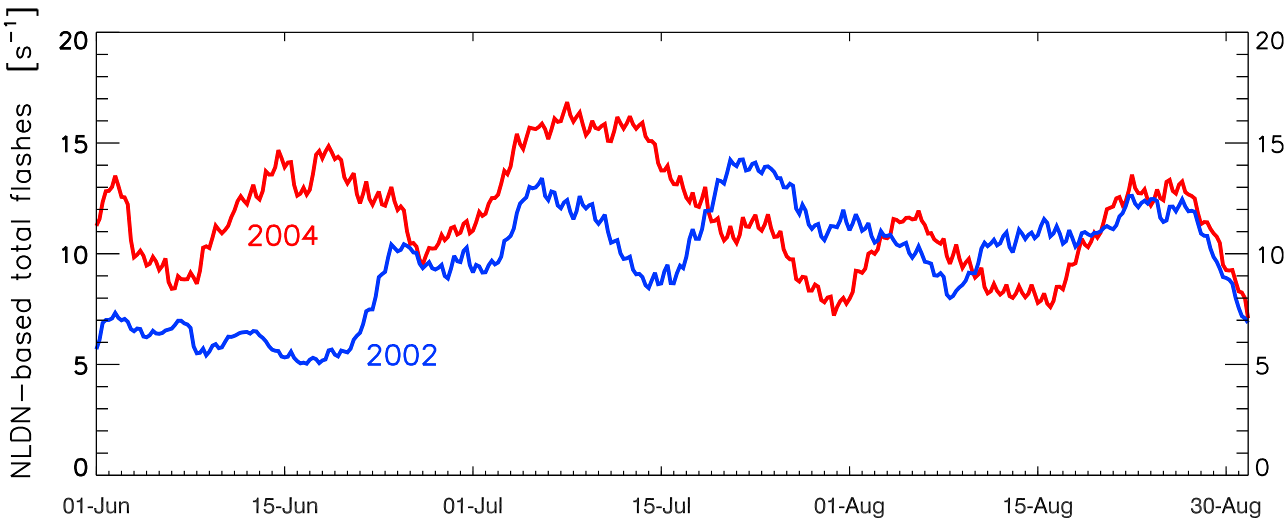

Figure 1. Total (IC + CG) lightning flash rates for the summers 2002 (blue) and 2004 (red) over the CONUS derived from the NLDN-observed CG flashes (adjusted by the IC/CG ratios). Flash rates are smoothed with a 7 day moving average.

meteorology [*Godowitch et al.*, 2008]. To quantify the impact of changes in meteorology and associated lightning, we use the UMD-CTM to simulate the summers of 2002 and 2004. Because of the wide availability of observations (INTEX-A), we use the summer 2004 as a reference year to evaluate the model performance with regard to lightning and implementation of pollution controls (NO_x SIP Call). We conduct three lightning simulations L0, L1 and L2 (Table 1) to account for current uncertainty in the simulation of lightning NO emissions and its relative role in the LRT of trace gases with respect to anthropogenic emissions.

3.1. Differences Between Summers 2002 and 2004

[30] Large summertime flash rates over the CONUS enhance the NA UT and outflow region with NO_x. Figure 1 shows the time series of NLDN-based total lightning over the CONUS in summer 2002 and 2004. Because of numerous thunderstorms in early summer 2004 (1 June to 17 July), lightning flash rates over the CONUS were about 50% higher compared to early summer 2002. In late summer (18 July to 31 August), total flash rates over the CONUS in 2004 were similar to those in 2002. Additionally, the onset of the North American Monsoon over the southwestern United States and northwestern Mexico for both years occurred in mid-July [*Li et al.*, 2004; *Gao et al.*, 2007]. Finally, there were contrasting patterns of vertical transport in early and late summer in the BL (Figure 2). In early summer, there was 10%–40% more convective lofting in 2004 than in 2002 over the ORV and much of the eastern, central and southern United States, with less lofting over New England. In late summer, there was less lofting over the central and southern United States in 2004, with more lofting over New England. Therefore, we break our analysis into two periods: early summer (1 June to 17 July) and late summer (18 July to 31 August).

[31] Figure 3 shows GEOS-4 surface temperatures and winds at ~5.5 km above the local surface. A prominent feature of the circulation over the United States is the strong low-level jet transporting air and moisture from the Gulf of Mexico to the central United States up to ~45°N (not shown). At ~5.5 km, a strong anticyclone dominates the south-central and southwestern United States, consistent with 4 year climatology shown in *Li et al.* [2005]. The anticyclonic circulation has important implications for the fate of convective outflow over the United States, as we discuss later. In addition to the upper level anticyclone, the northward expansion of the subtropical Bermuda High in the late summer influences the winds along the east coast of the United States In 2004, especially during early summer, enhanced westerlies over the eastern United States (Figure 3b), in combination with enhanced BL lofting (Figure 2b), promoted outflow of anthropogenic pollution from North America.

[32] Figure 4 shows the spatial pattern of NLDN-based IC + CG flashes during early summer 2002 and 2004. Higher flash rates were detected over most of the U.S in early summer 2004 compared to 2002: a factor of 2–4 increase over the Plains (Colorado, Nebraska, Kansas, Oklahoma and northern Texas) and a factor of 1.5–2 increase over the southern United States and parts of the ORV (southern Illinois, Indiana and Kentucky). The mean IC + CG flash rates over the CONUS were 8.30 flash s^{-1} and 12.94 flash s^{-1} in early summer 2002 and 2004, respectively.

[33] In order to compare lightning flash rates (IC + CG) observed from space and detected from the NLDN, we construct the time series shown in Figure 5. This comparison presents the sums over the CONUS south of 35°N as derived from NLDN and LIS observations. Both the NLDN and LIS time series agree that June and July of 2004 had increased flash rates with respect to 2002 in this region. However, we find that more lightning was observed by the NLDN network (after adjustment by the IC/CG ratios) than by the LIS sensor during summers 2002–2005. Similarly, *Jourdain et al.* [2010] found that NLDN-based flash

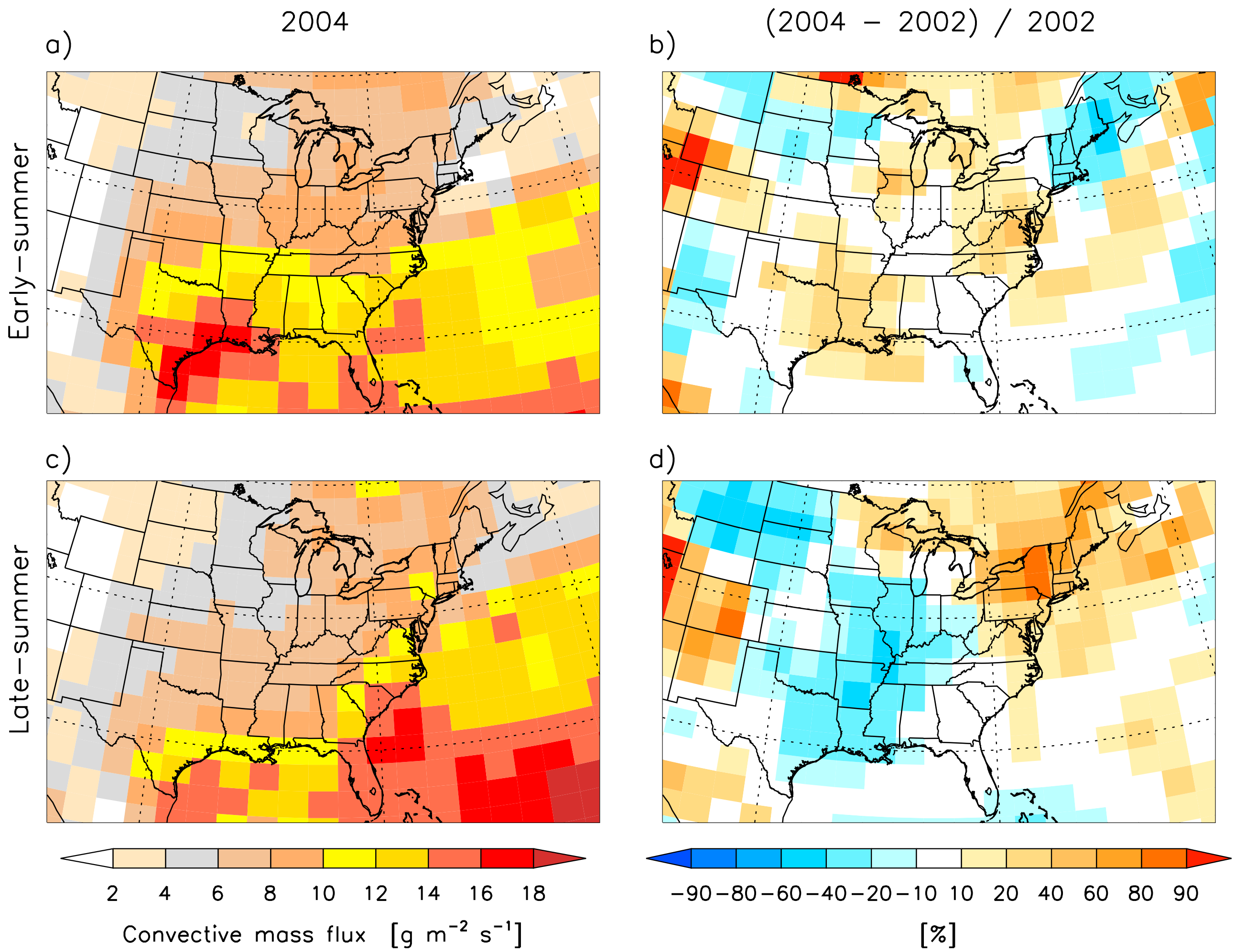

Figure 2. Convective mass fluxes (mean from the surface to 700 hPa) averaged for (a) 1 June to 17 July 2004 and (c) 18 July to 31 August 2004 and the relative change (%) between 2002 and 2004 averaged for (b) early and (d) late summer. Warm (cold) colors indicate more (less) vertical mixing by convection in 2004 than in 2002. Convective mass fluxes are calculated as the sum of deep convection and shallow convection fields from the GEOS-4 CERES reanalysis.

rates (assuming an IC/CG ratio of 3) over the CONUS (25°N–50°N) in July 2006 were about 40% higher than OTD/LIS flash rates. Over the CONUS south of 35°N, the summertime IC/CG ratios average 3.17 (when the grid boxes are weighted by the CG flash rates during 2002–2004). While we do not have a reason to believe that IC/CG ratios are overestimated, if we decrease this mean summertime IC/CG ratio from 3.17 to 1.41, then the mean combined flash rates (IC + CG) derived from the NLDN would be consistent with the ones derived from the LIS. In our analysis, we exclude weak positive flashes (peak current <20 kA) from the NLDN data. It should be noted that removing only 0–10 kA flashes, as done by *Boccippio et al.* [2001], would require the summertime IC/CG ratios in this region to be decreased even more (to IC/CG = 1.24) for an agreement between NLDN- and OTD/LIS-based estimates of total flash rate. Therefore, model flash rates from simulation L1 (adjusted to NLDN data) exceed model flash rates from simulation L0 (adjusted to OTD/LIS) as shown in the auxiliary material (Figure S3) for early summer 2004.

[34] To summarize, the LIS-derived flash rates are nearly a factor of 2 lower than NLDN-based IC + CG flash rates south of 35°N, suggesting either (1) a fraction of NLDN flashes with negative peak currents are actually IC flashes, (2) the climatological IC/CG ratios are overestimated, or (3) LIS flash rates are underestimated. The latter two possibilities could be caused by uncertainties resulting from temporal and spatial undersampling by LIS [*Boccippio et al.*, 2001]. The uncertainties of lightning detection by LIS are discussed by *Boccippio et al.* [2002].

3.2. UMD-CTM Comparison With Observations

3.2.1. Comparison With DC-8 in Situ Measurements During INTEX-A

[35] The INTEX-A field mission was conducted in summer 2004 (1 July to 15 August 2004) and focused on quantifying and characterizing the summertime inflow and outflow of pollution over North America and the western Atlantic [*Singh et al.*, 2006]. INTEX-A was an important

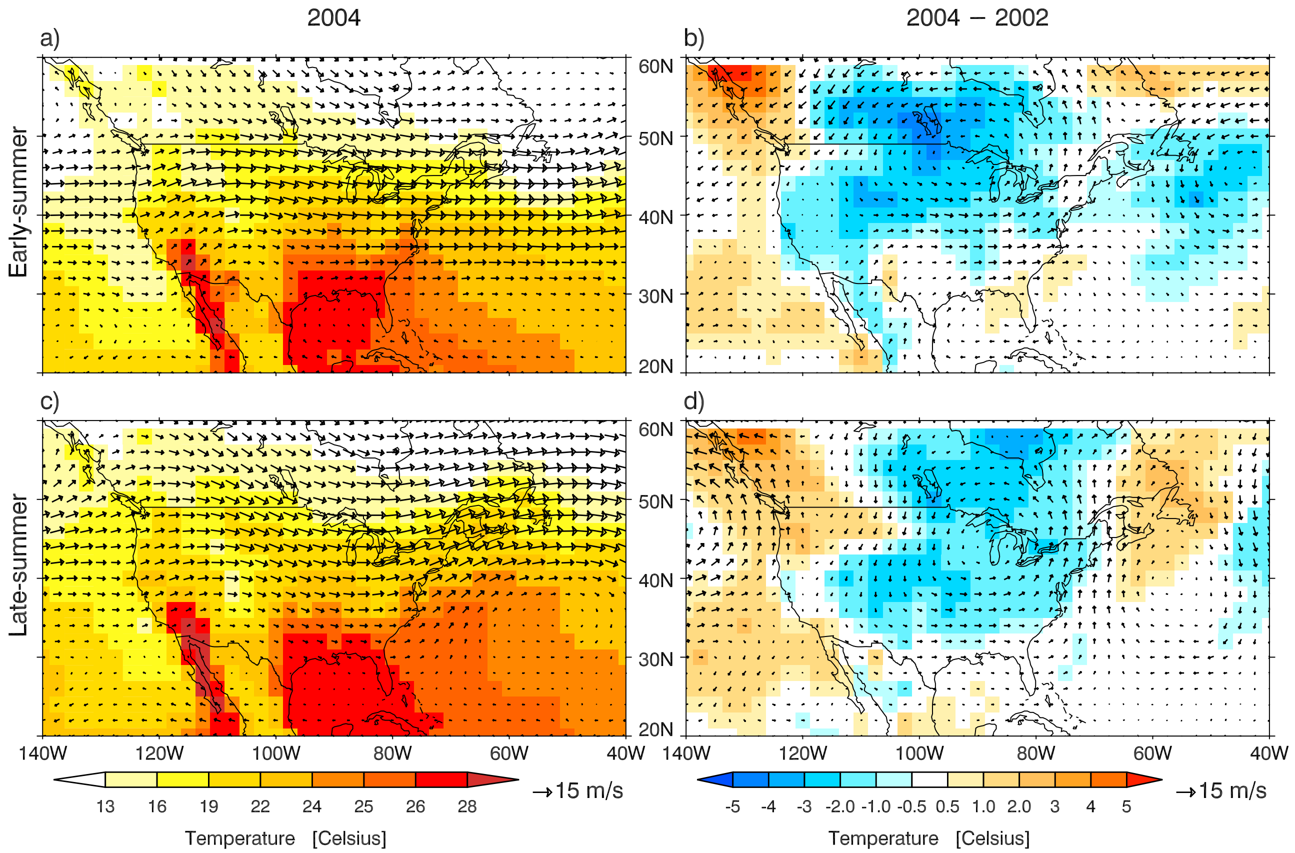

Figure 3. Mean GEOS-4 CERES surface temperatures and winds at ~5.5 km above the local surface for the period of (a) 1 June to 17 July 2004 and (c) 18 July to 31 August 2004 and the differences relative to 2002 for (b) early and (d) late summer.

component of the coordinated multiplatform atmospheric chemistry field program called ICARTT [*Fehsenfeld et al.*, 2006]. Here we use observations from NASA's DC-8 aircraft.

[36] Regional lightning is the dominant source of UT NO_x and can lead to O_3 increases of 10 ppbv or more in the UT [e.g., *DeCaria et al.*, 2005]. Deep convection and lightning were important factors during INTEX-A [*Hudman et al.*,

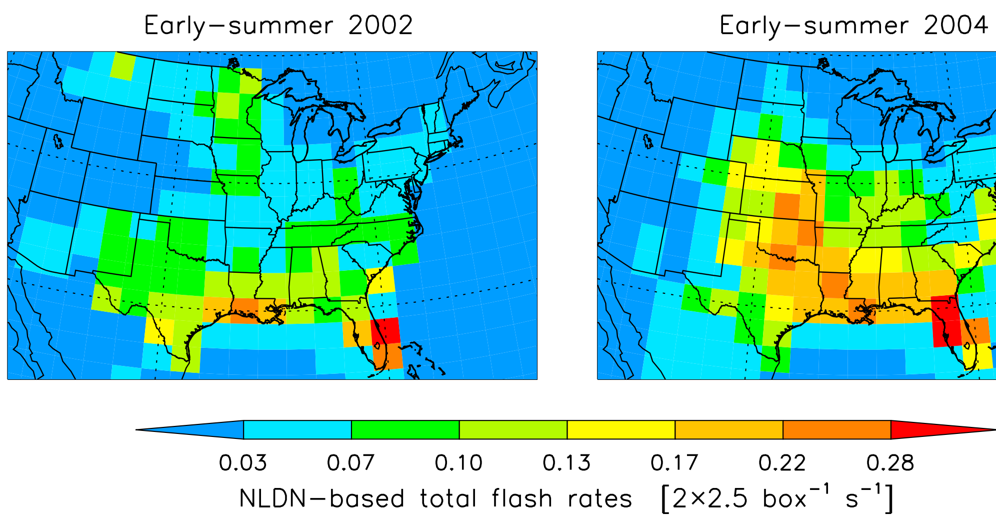

Figure 4. The NLDN-based total (IC + CG) lightning flash rates from (left) 2002 and (right) 2004 over the CONUS during early summer (1 June to 17 July). For comparison with the UMD-CTM flash rates for early summer 2004, see Figure S3 in the auxiliary material.

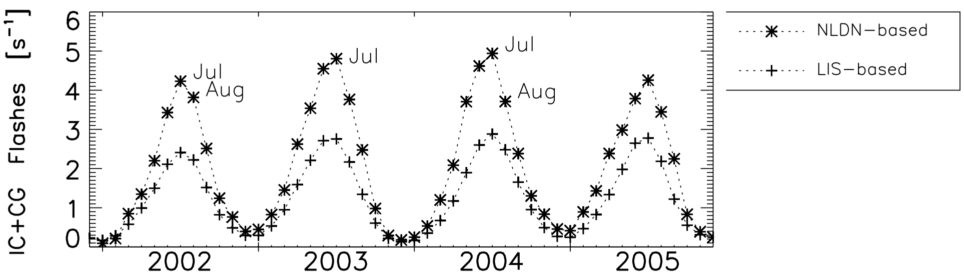

Figure 5. Total (IC + CG) lightning flash rates derived from NLDN ground and LIS spaceborne observations over the CONUS (land only) south of 35°N during 2002–2005. CG flash rates detected by the NLDN network (adjusted by the IC/CG ratios) are smoothed spatially (7.5° boxcar) and temporally (98 day window) and averaged for each month (indicated by the asterisks). Monthly LIS observations (LRMTS) were also smoothed with 98 day and 7.5° moving average.

2007; *Bousserez et al.*, 2007; *Singh et al.*, 2007]. Backward trajectories [*Fuelberg et al.*, 2007] indicated that the DC-8 often sampled lightning-influenced air, which makes summer 2004 an ideal test bed for the lightning parameterization schemes.

[37] We use aircraft (DC-8) observations from all INTEX-A flights (with the exception of Flight 3 over the eastern Pacific) to determine the model biases for the three lightning simulations L0, L1 and L2. The individual flight track profiles are mostly stair step ascents and descents covering large horizontal regions (details on flight paths are available in work by *Singh et al.* [2006]). Approximately half of the DC-8 samples were taken above 500 hPa (i.e., pressures <500 hPa). We compare simulated vertical profiles output hourly from the UMD-CTM to 1 min merge aircraft measurements with a focus on the UT model biases. Following the correction reported by *Ren et al.* [2008], observed OH values are scaled up by a factor of 1.64. To ensure a regionally representative signal, we remove biomass burning plumes (HCN > 500 pptv, CH_3CN > 225 pptv, or CO > 99th percentile), stratospheric air (O_3/CO > 1.25 mol mol^{-1}) and fresh pollution plumes (NO_x/NO_y > 0.4 mol mol^{-1}, or if NO_y is not available, NO_2 > 4 ppbv and height <3 km) from observations [*Hudman et al.*, 2007]. This filtering excludes 8% (fresh pollution), 7% (biomass burning) and 3% (stratospheric air) of the DC-8 data, including some samples with a strong lightning NO signal. While the stratospheric filter only removes 3% of samples, it has the largest impact on UT O_3 biases. We sample the UMD-CTM at the locations and times of DC-8 filtered measurements using nearest neighbor values and interpolating to DC-8 heights.

[38] Figure 6 compares simulated and measured mean vertical distributions of NO, O_3, CO, HNO_3, NO_x, OH, PAN and NO_y. First, we analyze the UMD-CTM biases from the standard simulation (L1). Simulated NO and NO_x profiles are C shaped, reflecting the partitioning of LNO_x in the vertical [*Pickering et al.*, 1998] and the anthropogenic source near the surface. Biases are largest in the UT. NO is underestimated throughout the column (30%–60% too low at 500–300 hPa and 80% too low above 300 hPa). NO_x is underestimated by 20%–50% at 500–300 hPa and by 80% above 300 hPa. The simulation with doubled lightning NO production per flash (L2) decreases biases for both NO and NO_x to 10%–30% at 500–300 hPa and to 60% above 300 hPa; NO_x agrees well with measurements below 300 hPa.

[39] O_3 is overestimated in the LT by 15 ppbv, but this bias drops to 9.6 ppbv above 500 hPa. Doubling the lightning NO source (simulation L2) increases the bias to 12.3 ppbv. A 2–3 ppbv increase in UT O_3 resulting from a doubling of the LNO_x source is also seen at IONS sites (Figure S4 in the auxiliary material). The largest impact is seen near (Houston, Texas, and Huntsville, Alabama) and downwind (Wallops Island, Virginia, and Sable Island, Nova Scotia) of frequent thunderstorms. The UMD-CTM also shows a considerable low bias of 20% for CO throughout the column compared to aircraft measurements. CO is not sensitive to different LNO_x sources. Upper tropospheric OH is highly sensitive to LNO_x, which can be seen in the clear separation of the L0, L1 and L2 profiles. Observations indicate that OH concentrations increase with altitude; this slope is best captured in the L2 simulation. Mean absolute values of bias above 500 hPa are 0.11 pptv and 0.07 pptv for L1 and L2, respectively.

[40] HNO_3 during INTEX-A was measured by the California Institute of Technology (CIT) and the University of New Hampshire (UNH). HNO_3 is often depleted in the free troposphere because of scavenging during convection but can increase downwind of convection due to oxidation of NO_2. HNO_3 is highly sensitive to lightning; we see a larger change from L1 to L2 simulated profile than for PAN, consistent with *Hudman et al.* [2007] and *Labrador et al.* [2005]. Variability of HNO_3 in the LT is larger than in the UT and is associated with variability of NO_x. The simulated HNO_3 is generally overestimated with respect to both the CIT (Figure 6) and UNH (not shown) data sets. The CIT observed about 40% more HNO_3 above 500 hPa than the UNH, leading to better agreement with the model. HNO_3 is overestimated likely because of NO_x oxidation being too rapid or wet removal being insufficient in the model or both. Overall, despite NO_x underprediction, NO_y is overpredicted below 300 hPa because both PAN and HNO_3 are overestimated with respect to the in situ measurements. Most of NO_y high bias above 500 hPa is due to PAN overestimation (by up to 0.2 ppbv and 0.3 ppbv in the L1 and L2 simulations, respectively). NO_x underestimation and NO_y overestimation could indicate a fundamental problem with the UT NO_y chemistry. *Henderson et al.* [2010] evaluated seven different chemical mechanisms. They found that each mechanism overestimates the rate at which NO_x is converted to NO_z ($NO_y - NO_x$), i.e., the rate at which NO_x ages.

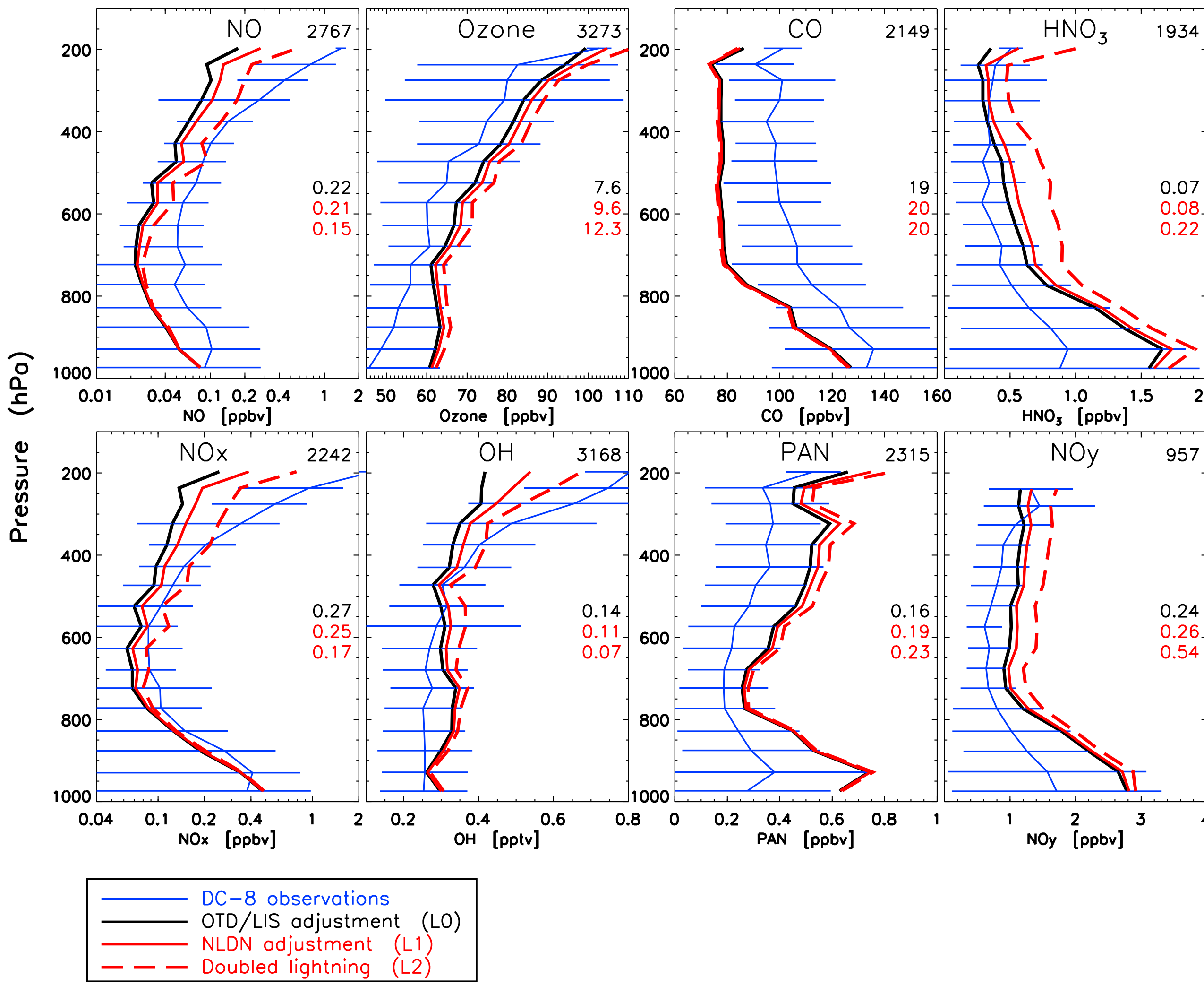

Figure 6. Mean vertical profiles of NO, O_3, CO, HNO_3, NO_x, OH, PAN, and NO_y. Measurements from the DC-8 aircraft (solid blue) are compared to UMD-CTM results from the L0 (solid black), L1 (solid red), and L2 (dashed red) simulations. The 1 min average measurements are compared to hourly UMD-CTM output sampled along the flight tracks. NO_y is estimated as the sum of its main oxidation products (NO_x, PAN, and HNO_3). We use here HNO_3 data obtained by a science team from the California Institute of Technology. Horizontal bars indicate standard deviations on the observations in each 50 hPa bin. The number of filtered observations above 500 hPa is listed in the top right corner of each plot. The absolute values of the bias (model minus observation) averaged from 50 hPa bins above 500 hPa are listed in the middle right part of each plot: L0 bias (top value), L1 bias (center value), and L2 bias (bottom value). Note the logarithmic scale for NO and NO_x.

They also suggested several updates and fixes to various mechanisms to slow down this conversion rate.

[41] In summary, doubling the lightning NO production per flash reduces NO, NO_x and OH biases in the UT. However, it increases the biases for O_3 and PAN slightly (by factors of 1.3 and 1.2, respectively) and for HNO_3 and NO_y substantially (factors of 2.8 and 2.0, respectively). In spite of increased O_3 biases (by 2.7 ppbv above 500 hPa, L1 versus L2), the NO_x profile from the L2 simulation agrees well with DC-8 measurements below 300 hPa but is low biased by 60% above 300 hPa. Because the lightning sources in L1 and L2 simulations bracket the emissions of other investigators (Table 2) and because of the mixed results from comparison with aircraft measurements during INTEX-A (NO_x underestimated; NO_y and O_3 overestimated) as to which lightning source is most realistic, we complement the L1 results by examining the impact of doubled lightning NO production per flash (L2).

3.2.2. Comparison With SCIAMACHY Measurements

[42] The impact of reduced NO_x emissions on NO_2 columns in the ORV (the region dominated by power plants that had implemented controls) is evident from space [*Kim et al.*, 2006]. Data from the high-resolution (30 × 60 km^2) SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) instrument, onboard ENVISAT, became available in August 2002. It detects the sunlight reflected from the Earth or scattered in the atmosphere [*Richter et al.*, 2005]. In nadir mode, SCIAMACHY

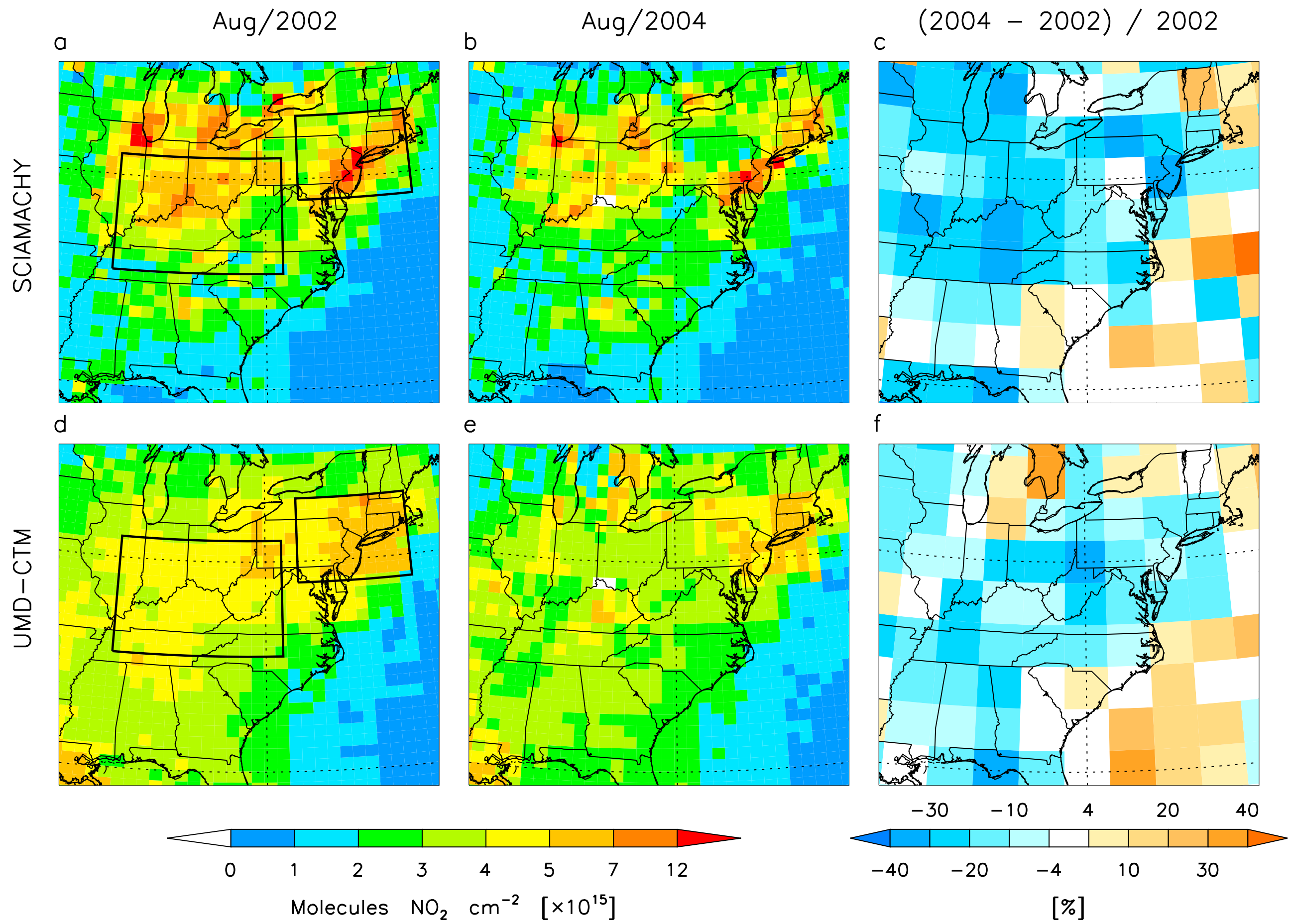

Figure 7. Comparison between (a, b) SCIAMACHY-observed and (d, e) UMD-CTM-simulated mean tropospheric NO_2 vertical columns (10^{15} molecules NO_2 cm^{-2}) composited from all overpasses with cloud fraction <0.3 over the eastern United States in (left) August 2002 and (middle) August 2004. The UMD-CTM is sampled at each SCIAMACHY pixel. Both observed (Figures 7a and 7b) and modeled (Figures 7d and 7e) columns are composited to a 0.54° × 0.70° (about 60 × 60 km^2) grid which is close to the native resolution of SCIAMACHY. In the ORV region (indicated by the larger box on the left), SCIAMACHY-observed reduction from 4.27 to 3.32 on average between 2002 and 2004, whereas simulated columns were reduced from 4.24 to 3.43 (all values in units of 10^{15} NO_2 molecules cm^{-2}). (c, f) Relative change (%) between 2002 and 2004 but at the 2° × 2.5° resolution.

observes the total NO_2 column from which the stratospheric column, as derived over the Pacific sector, is subtracted to get the tropospheric column. The measurements were screened to use only pixels with less than 30% cloud fraction.

[43] In Figure 7, we determine whether changes in satellite-observed and model-calculated NO_2 columns (L1 simulation) are consistent with the updated emission inventories. We sample the UMD-CTM by following daily satellite tracks (ENVISAT overpasses the ORV at 0930–1000 local standard time). The UMD-CTM tropospheric NO_2 columns are calculated by integrating the columns from the surface to the GEOS-4 CERES tropopause. The UMD-CTM columns are interpolated in space and time to the SCIAMACHY pixels and times using the outputs at 1200 and 1800 UTC. We composite the observed and simulated columns onto a 0.54° × 0.70° grid, corresponding to approximately 60 × 60 km^2 (Figures 7a, 7b, 7d, and 7e). By temporally averaging the UMD-CTM output at this resolution (each grid box includes ~8 clear-sky days during August 2002 and ~6 clear-sky days during August 2004) we are able to obtain more detail than can be obtained from 2° × 2.5° UMD-CTM. However, the UMD-CTM even when averaged in this fashion is unable to resolve the maxima observed by SCIAMACHY over polluted urban areas (e.g., Chicago).

[44] The observations from SCIAMACHY show that major NO_2 plumes over the northeastern United States are of substantially smaller magnitude in 2004 than 2002, consistent with the emission reductions due to the NO_x SIP Call. In August 2002, nearly one third of U.S. power plant NO_x emissions were from the ORV region (larger box in Figure 7a), while in August 2004 they comprised less than one fifth due to implemented emission controls. CEMS data show that power plant NO_x emissions from this region were reduced by ~50% between August 2002 and 2004. The observed NO_2 columns over the ORV were reduced by 22% on average from 2002 to 2004. A smaller observed reduction is expected because of transport from regions where emis-

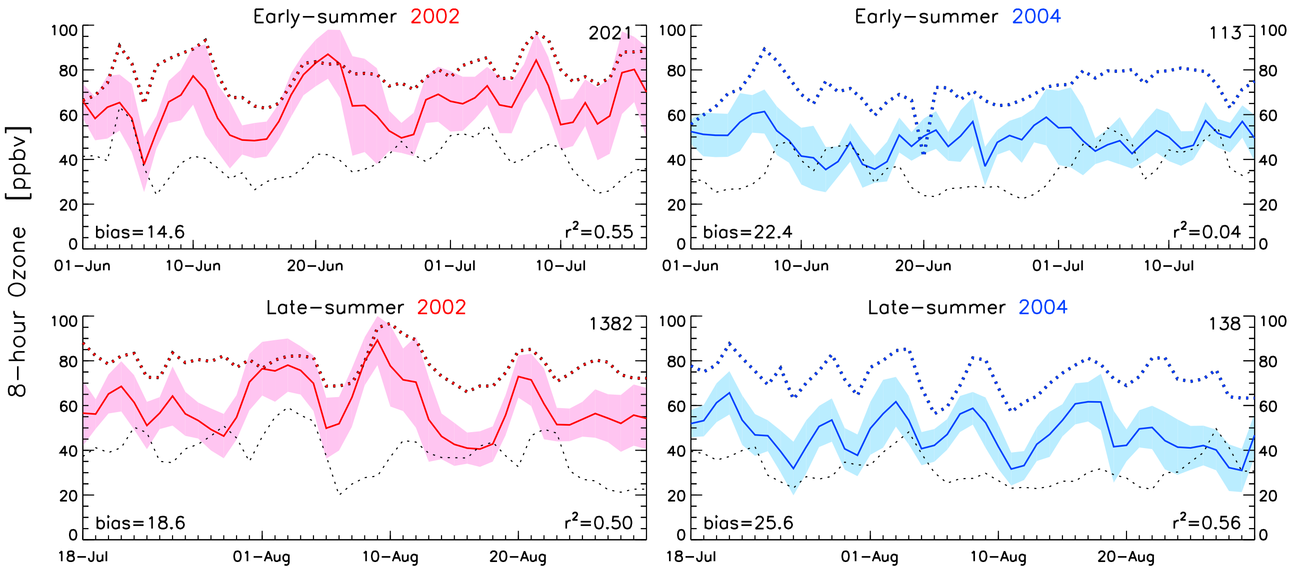

Figure 8. The 8 h O_3 time series constructed from 155 AQS sites located in the ORV region (indicated by the larger box in Figure 7a) in summer (left) 2002 and (right) 2004. The shaded area around the mean AQS-measured values (red and blue solid lines) indicates the standard deviation. The number of current NAAQS exceedances (8 h O_3 > 75 ppbv) for each month is in the top right corner of each plot. The UMD-CTM (dotted lines) is sampled at the locations of the AQS sites. The 8 h O_3 from the sensitivity simulation with the anthropogenic emissions from North America turned off (Table 1) is indicated by black dotted lines. The UMD-CTM bias is listed in the bottom left corner of each plot. The explained variance r^2 between time series of observed and simulated O_3 is in the bottom right corner of each plot.

sions reductions were smaller and because SCIAMACHY detects all types of NO_2, not just that resulting from power plant emissions. Similar reductions of 19% are seen in the UMD-CTM (Figures 7d and 7e). Reductions in observed columns were also seen over the northeastern United States (smaller box in Figure 7a), which is downwind of the ORV power plants. The modeled reductions in this region were 11%, while observed reductions were 20%. The difference in reductions between SCIAMACHY and the model in this region could indicate that NO_x emissions from other sources also decreased between 2002 and 2004; however, this difference may also indicate that the medium-range transport of NO_2 is underpredicted by the UMD-CTM. *Gilliland et al.* [2008] noted that CMAQ underestimated the response of O_3 to changes in emissions partly because the transport of O_3 and its precursors was underestimated. *Godowitch et al.* [2008] in a simulation with CMAQ also underestimated the improvement in air quality between summers 2002 and 2004. In 2002, there was a 2 ppbv low bias whereas 2004 had a 4 ppbv high bias in 8 h O_3.

3.2.3. Comparison With Observations From Ground-Based AQS Sites

[45] The AQS (Air Quality System) is an EPA database (http://www.epa.gov/ttn/airs/airsaqs) that provides ambient concentrations of air pollutants at monitoring sites, primarily in cities and towns. We use daily maximum 8 h O_3 (8 h O_3) as the metric for our comparisons, since it is important from a regulatory perspective. The 2008 National Ambient Air Quality Standard (NAAQS) for 8 h O_3 is 75 ppbv.

[46] Figure 8 shows time series of 8 h O_3 from the ORV region. We see that the UMD-CTM captures well the periods with large day-to-day variability. Most of the variance (r^2 > 0.5) in summer 2002 and in late summer 2004 is explained by the model. In early summer 2004, observed day-to-day variability is smaller, and the UMD-CTM is unable to capture these subtle changes in surface O_3. It is likely that the correlation between simulated and observed 8 h O_3 might be improved if daily CEMS data were used in the model instead of monthly CEMS data. In the ORV, 8 h O_3 is simulated with mean high biases of 16.5 ppbv and 24.0 ppbv in summer 2002 and 2004, respectively. The difference between these biases is likely to decrease by up to 3 ppbv (see section 2.3) if isoprene emissions are allowed to respond to temperature variations. Despite this mean high bias, O_3 concentrations at numerous AQS sites were slightly underpredicted during the highest O_3 episodes that occurred in summer 2002. Similarly, in the rest of the eastern United States (not shown), the UMD-CTM captures the O_3 variations reasonably well (r^2 > 0.5) with biases of 10–20 ppbv seen over the Great Lakes (Wisconsin and Michigan), New York State and New England and with 20–30 ppbv biases seen in the southern United States. Overprediction of surface O_3 in the eastern United States during summer is common in many CTMs. *Reidmiller et al.* [2009], as part of the Hemispheric Transport of Air Pollution project, 2007 (http://htap.org), showed that the mean multimodel (16 CTMs) bias was 10–20 ppbv. Some of the bias is due to the spatial averaging of emissions in a model grid box; dilution of NO_x emissions to a 2° × 2.5° grid leads to greater O_3 production than at finer resolutions [*Sillman et al.*, 1990; *Park et al.*, 2004b]. Additional uncertainty is introduced by the treatment of isoprene emissions (BEIS versus MEGAN) and isoprene-nitrate chemistry in chemical mechanisms.

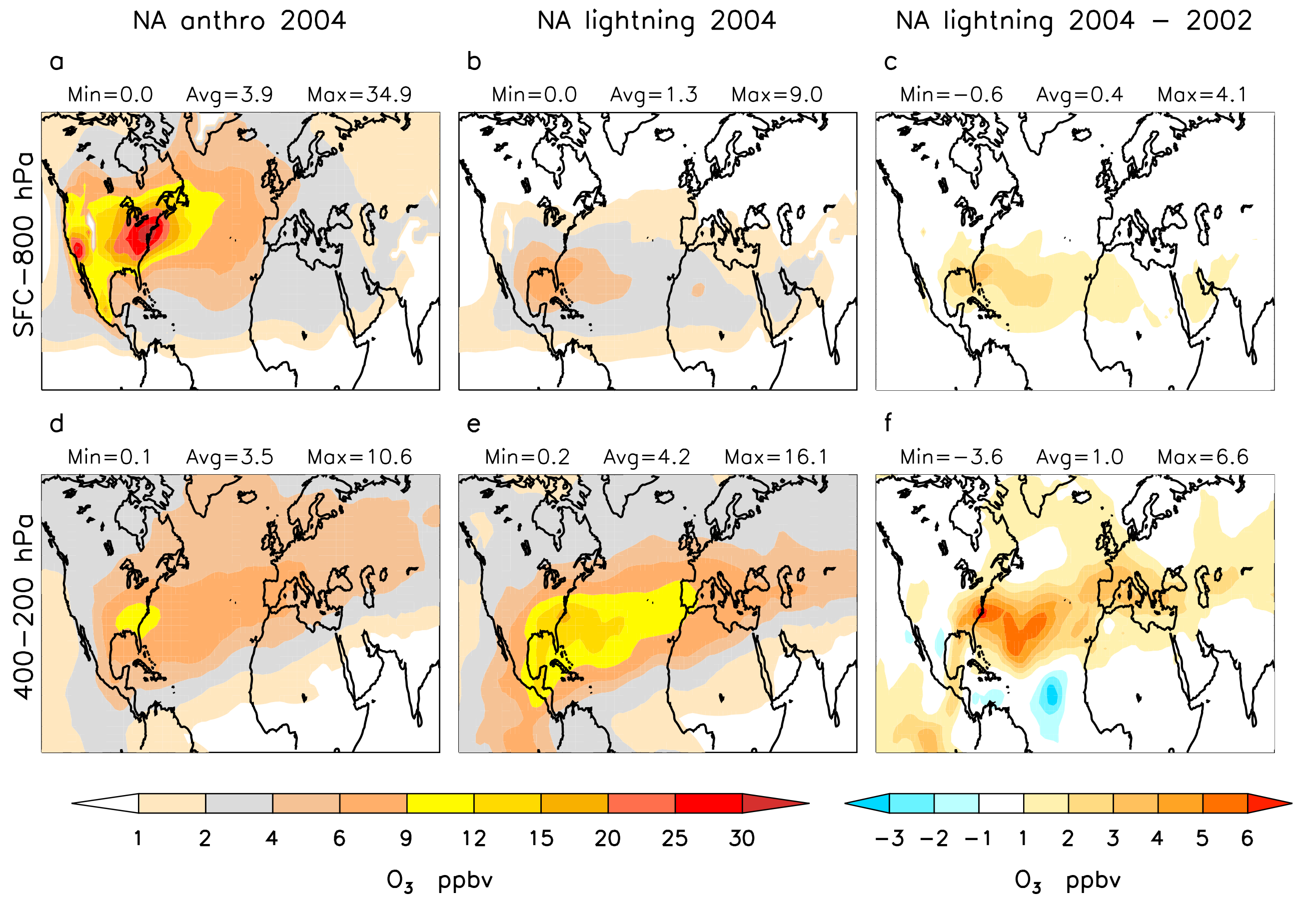

Figure 9. Early summer (1 June to 17 July) O_3 enhancements from North American (a, d) anthropogenic and (b, e) lightning emissions as diagnosed by simulation L1 and the sensitivity simulations with the respective sources turned off (Table 1). (c, f) Difference between 2004 and 2002 due to NA lightning. The values are averaged in (top) the lower troposphere (from the surface to 800 hPa) and (bottom) the upper troposphere (400–200 hPa). Minima, averages, and maxima are listed above each plot.

[47] Overall, the combined effects of changes in meteorology and emissions had a great impact on 8 h O_3 concentrations in the northeastern United States. Observed 8 h O_3 concentrations were reduced by 14 ppbv in the ORV on average from summer 2002 to 2004. Reductions of 7 ppbv were seen in the UMD-CTM (sampled at the locations of the AQS sites).

3.3. Ozone Enhancements From NA Anthropogenic Emissions and Lightning

[48] We compare the relative effects of modeled O_3 enhancements from NA anthropogenic emissions and NA lightning for summer 2004, focusing on long-range transport and continental outflow. Although not strictly true, because of the nonlinear response of O_3 to NO_x emissions, the lightning (anthropogenic) enhancement is diagnosed as the difference between simulations with and without the LNO_x (anthropogenic) source. These results are then compared to similar simulations for summer 2002, allowing us to quantify the impact of changes in meteorology and NA lightning between summers 2002 and 2004 on long-range transport and continental outflow as well as the relative radiative impact.

[49] Figures 9a, 9b, 9d, and 9e show the O_3 enhancements from NA anthropogenic emissions and lightning in early summer 2004. Anthropogenic emissions produced the greatest O_3 enhancements near the surface (up to 35 ppbv over the eastern United States, as seen in Figure 9a), whereas lightning had the greatest impact in the UT (up to 16 ppbv over the Gulf Coast and the western North Atlantic, as seen in Figure 9e). *Hudman et al.* [2009], using the GEOS-Chem, compared the O_3 enhancements from NA anthropogenic emissions and lightning during INTEX-A. They also showed that the BL enhancements were mainly anthropogenic and that lightning had the greatest impact in the UT. Convectively lifted precursors and LNO_x enhanced the O_3 production, especially at higher altitudes. We see similar enhancements as *Hudman et al.* [2009] over North America and in the continental outflow, near-surface enhancements from anthropogenic emissions of 3–5 ppbv over western Europe, and UT (400–200 hPa) enhancements from lightning of 9–12 ppbv over the eastern subtropical Atlantic (with 9 ppbv contour reaching over Spain). In contrast with *Hudman et al.* [2009], we also examine the 2002 to 2004 variations in O_3 enhancements from lightning (Figures 9c and 9f). From 2002 to 2004, there is an increase in the

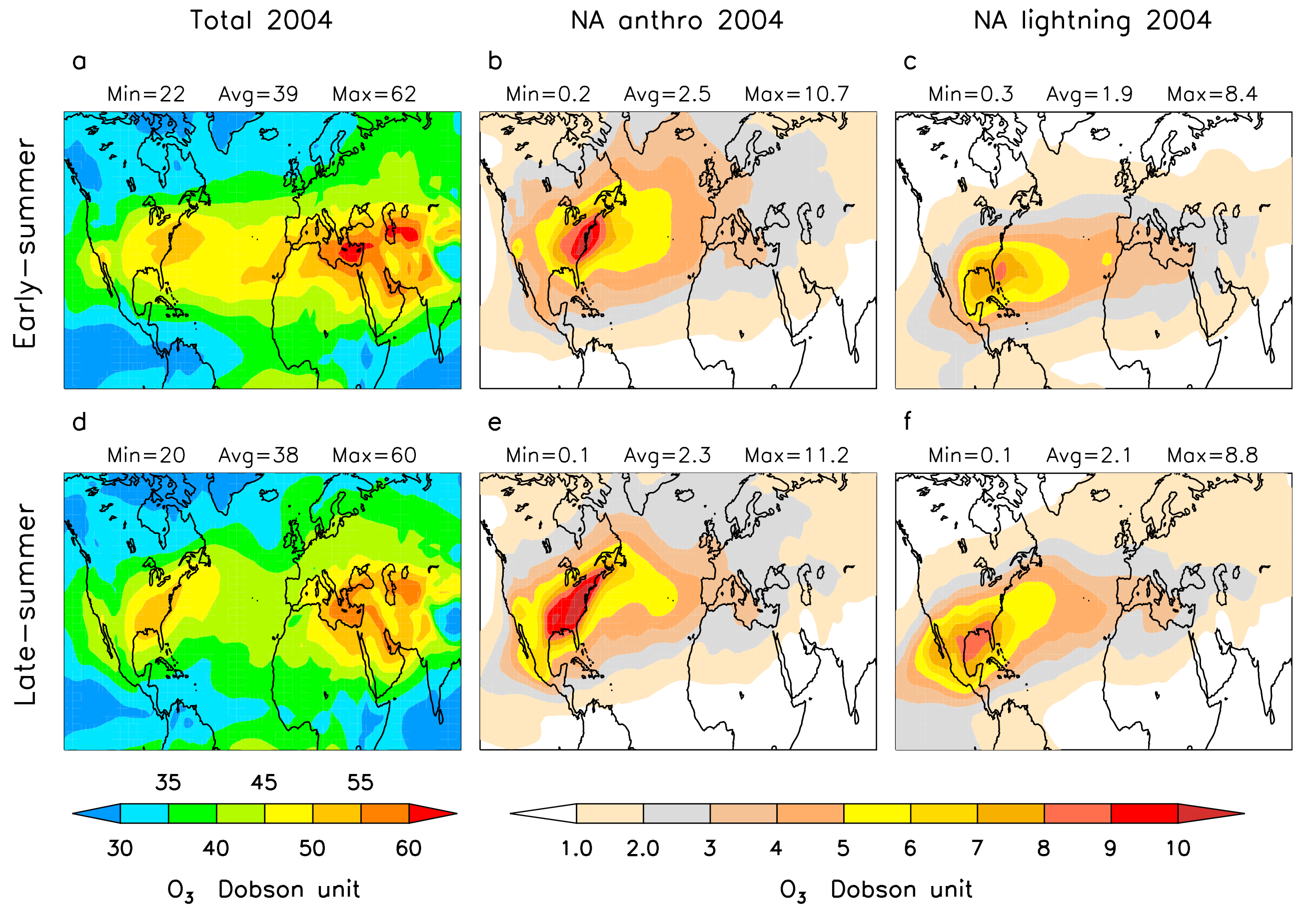

Figure 10. (a, d) Tropospheric O_3 columns and their enhancements due to North American (b, e) anthropogenic emissions and (c, f) lightning as diagnosed by simulation L1 and the sensitivity simulations with respective sources turned off (Table 1). The values are (top) early summer (1 June to 17 July) mean and (bottom) late summer (18 July to 31 August) mean for 2004. Minima, averages, and maxima are listed above each plot. Corresponding NO_2 columns are in Figures S5a–S5c and S5e–S5g in the auxiliary material.

contribution of lightning to eastern United States. O_3, both near the surface (Figure 9c) and in the UT (Figure 9f).

[50] Figure 10 shows the tropospheric O_3 columns during early and late summer 2004 and the enhancements from anthropogenic emissions and lightning NO emissions. In late summer 2004, the warm conveyor belt was especially active [*Kiley and Fuelberg*, 2006], enhancing the transport of anthropogenic O_3 along the U.S. east coast. In addition, stronger southerly winds relative to early summer (Figure 3c versus Figure 3a) pushed the anthropogenic and lightning plumes to more northern latitudes. Maximum anthropogenic enhancements were ~11 DU, which represented ~21% of the total tropospheric column. Maximum lightning enhancements were ~9 DU (Figures 10c and 10f), which represented ~18% of the total tropospheric column. Doubling the LNO_x source over North America in the UMD-CTM produces an additional 4 ppbv of O_3 in the UT (400–200 hPa) compared to lightning enhancements from L1 simulation, thus adding ~2 DU to the tropospheric column. Doubled lightning (L2) produces therefore ~11 DU, which is the same as produced from anthropogenic emissions. The relatively small increase from ~9 to ~11 DU, when going from 240 NO mol flash^{-1} to 480 NO mol flash^{-1} is due to the nonlinear response of O_3 enhancements to LNO_x emissions. Similarly, in downwind regions over the northern Atlantic and western Europe, the early summer O_3 enhancements in the L2 simulation exceed the L1 enhancements by only 33% despite a doubling of LNO_x emissions from North America. In contrast, *Wu et al.* [2009] found no significant nonlinearity of O_3 response to ANO_x emissions in the downwind regions from North America.

[51] Shifting our focus to the summer-to-summer change, Figures 11a and 11c indicate substantial differences between 2004 and 2002 in O_3 enhancements due to lightning in the continental outflow, especially for early summer. Early summer 2004 lightning led to 2–6 ppbv more O_3 in the UT (400–200 hPa) over the subtropical North Atlantic, southern Europe and the Middle East when compared to 2002 (Figure 9f), enhancing the tropospheric column by 1.0–3.5 DU. Over the subtropical North Atlantic (20°N–45°N), increased NA lightning (and changes in meteorology) between early summer 2002 and 2004 explain about two thirds of the increase in tropospheric O_3 column between 2002 and 2004 (not shown). During late summer both years had similar lightning flash rates; however, meteorological conditions in 2004 were less favorable for O_3 formation [see

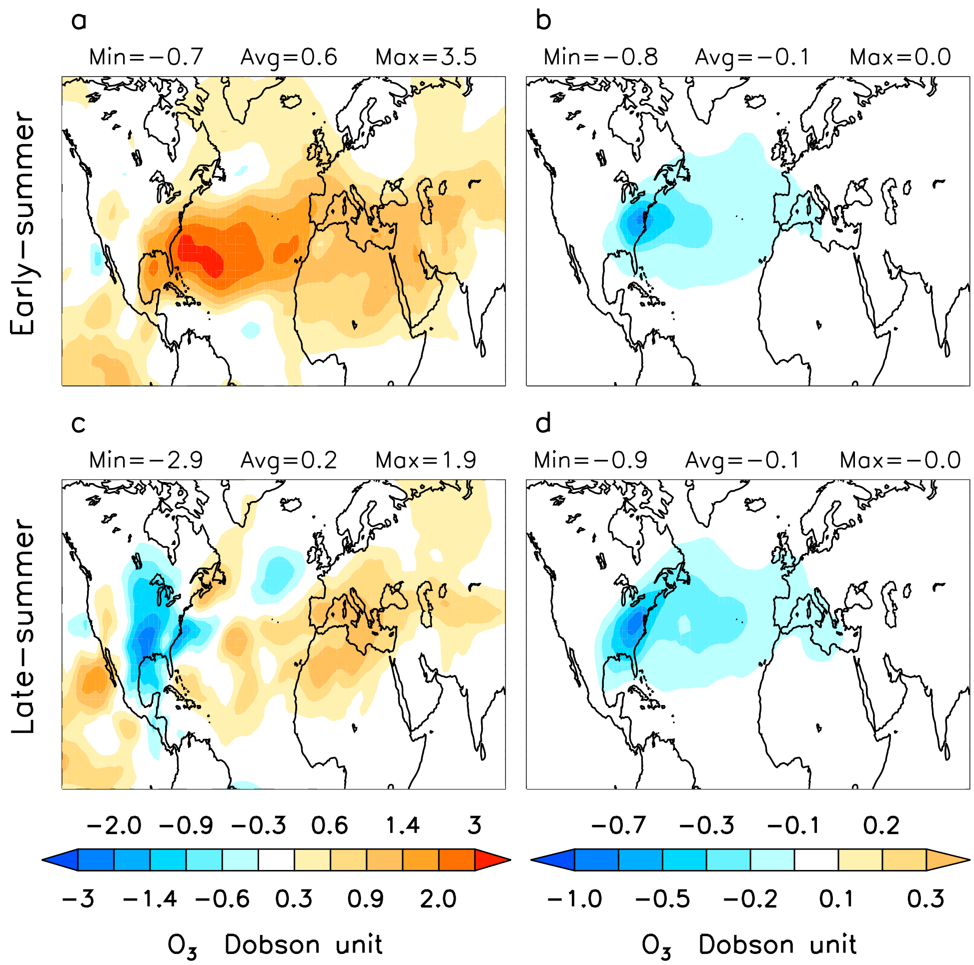

Figure 11. (left) The impact of changes (2004 minus 2002 difference) in North American lightning and meteorology and (right) the impact of NO_x SIP Call reductions on tropospheric O_3 columns as diagnosed by simulation L1 and three sensitivity simulations (Table 1), two with the LNO_x source turned off (one for each year) and one with 2002 power plant NO_x emissions and 2004 flash rates and meteorology. The values are (top) early summer (1 June to 17 July) mean and (bottom) late summer (18 July to 31 August) mean. Minima, averages, and maxima are listed above each plot.

also *Cooper et al.*, 2009; *Allen et al.*, 2010] and led to 1–4 ppbv less O_3 in the UT over the eastern United States than in 2002 (not shown). However, the tropospheric column over the eastern Atlantic, Europe and northern Africa had up to 1.2 DU more O_3 from lightning in 2004 than 2002 (Figure 11c), reflecting more efficient transport over the Atlantic in both early and late summer 2004. Later in the summer (both years), the LIS sensor observed enhanced flash rates over Mexico, indicating the increased lightning activity and deep convection associated with the North American Monsoon [*Li et al.*, 2004; *Gao et al.*, 2007]. Note the intensification of the O_3 enhancement over Mexico and the convective outflow over the Gulf of Mexico in late summer (Figure 10c versus Figure 10f).

[52] To compare the impact of increased NA lightning (and changes in meteorology) between 2002 and 2004 with the impact of emission reductions resulting from the NO_x SIP Call, we did a sensitivity simulation with 2002 NO_x emissions (when power plant emissions were not as tightly regulated) and 2004 meteorology and lightning. While decreases in surface layer O_3 over the eastern United States because of the NO_x SIP Call were substantial (not shown), decreases in the tropospheric column were small (Figures 11b and 11d). Columns were reduced by <0.5 DU over the North Atlantic in both early and late summer (with the −0.1 DU contour reaching Spain, Italy and southern France). During the early summer in the region where the emission reductions (between 2002 and 2004) had the largest impact on O_3 column over the North Atlantic (30°N–50°N), the change in O_3 column due to changes in lightning and meteorology exceeded the absolute value of the change due to reduced anthropogenic emissions by a factor of 7. Differences in flash rates between 2002 and 2004 were much smaller in late summer. Consequently, the changes in O_3 column due to changes in lightning and meteorology were also smaller, and the change in O_3 column due to changes in lightning and meteorology was comparable to the change due to reduced ANO_x emissions but opposite in sign.

[53] Lightning enhancements are primarily in the UT, where previous studies [e.g., *Lacis et al.*, 1990] have shown

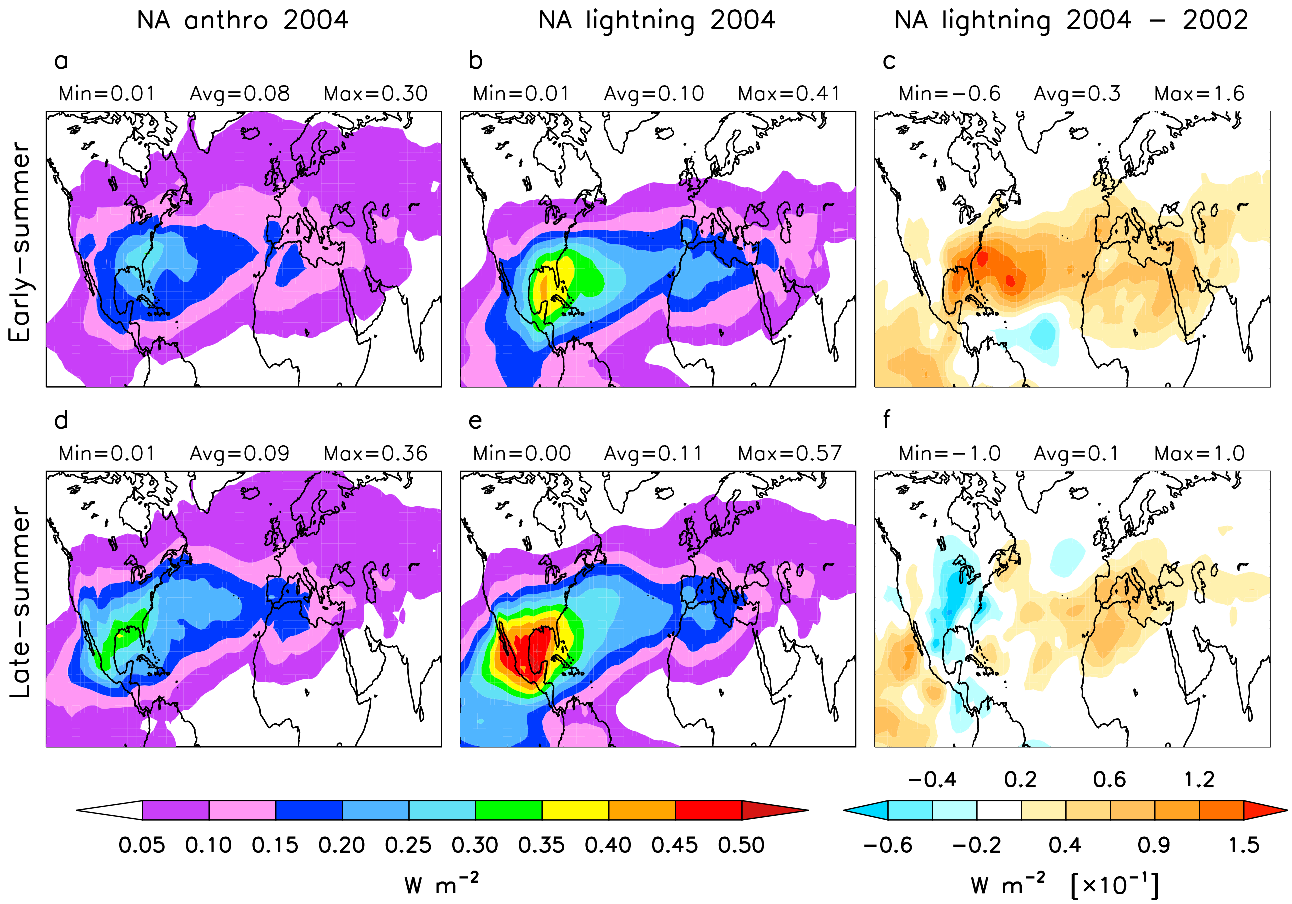

Figure 12. RF (calculated as described in section 2.5) for 2004 due to (a, d) North American anthropogenic emissions and (b, e) lightning as diagnosed by simulation L1 and the sensitivity simulations with respective sources turned off (Table 1). (c, f) Difference between 2004 and 2002 due to NA lightning. The values are (top) early summer (1 June to 17 July) mean and (bottom) late summer (18 July to 31 August) mean. Minima, averages, and maxima are listed above of each plot. Note a factor of 10 smaller units for RF in Figures 12c and 12f.

O_3 to be most effective as a greenhouse gas. *Choi et al.* [2009], using the regional chemistry transport model REAM for the period of June–July 2005, showed that in the immediate convective outflow, the radiative effects of O_3 produced from LNO_x were up to three times as large as those from anthropogenic emissions. In our analysis, we examine the larger-scale radiative impact (defined in section 2.5) due to O_3 produced from anthropogenic emissions (RF_{anthro}) and from lightning (RF_{LNOx}). Figure 12 shows that RF_{anthro} ranged from 0.15 to 0.30 W m^{-2} in the continental outflow across the North Atlantic, whereas the RF_{LNOx} ranged from 0.20 to 0.40 W m^{-2} (0.25–0.50 W m^{-2} for doubled LNO_x source) over the same area in early and late summer 2004. The RF_{LNOx} also exceeded RF_{anthro} over southern Europe and northern Africa in both early and late summer 2004. We find that, in early summer 2004, the RF_{LNOx}/RF_{anthro} ratio ranged from 0.3 at higher latitudes of the North Atlantic to ~1.6 over the subtropical North Atlantic, with a maximum of 2.3 over the southern Gulf of Mexico (Figure 13). Doubling the LNO_x source has the greatest impact on this ratio over the western subtropical North Atlantic, where the RF_{LNOx}/RF_{anthro} ratio increases from 1.6 to 2.1.

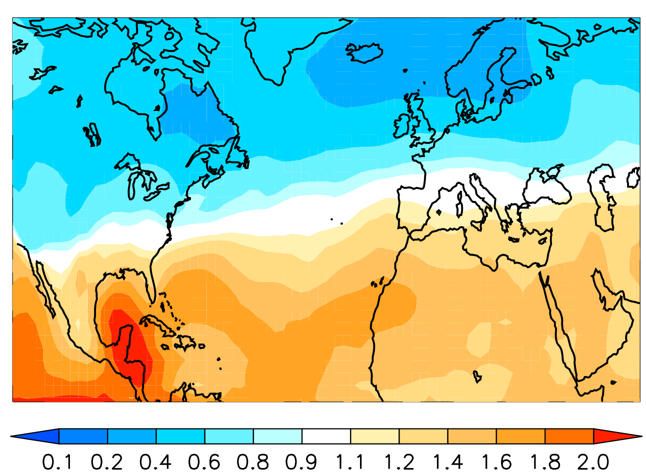

Figure 13. Early summer 2004 (1 June to 17 July) ratio RF_{LNOx}/RF_{anthro} as diagnosed by the standard simulation and sensitivity simulations with respective sources turned off (Table 1).

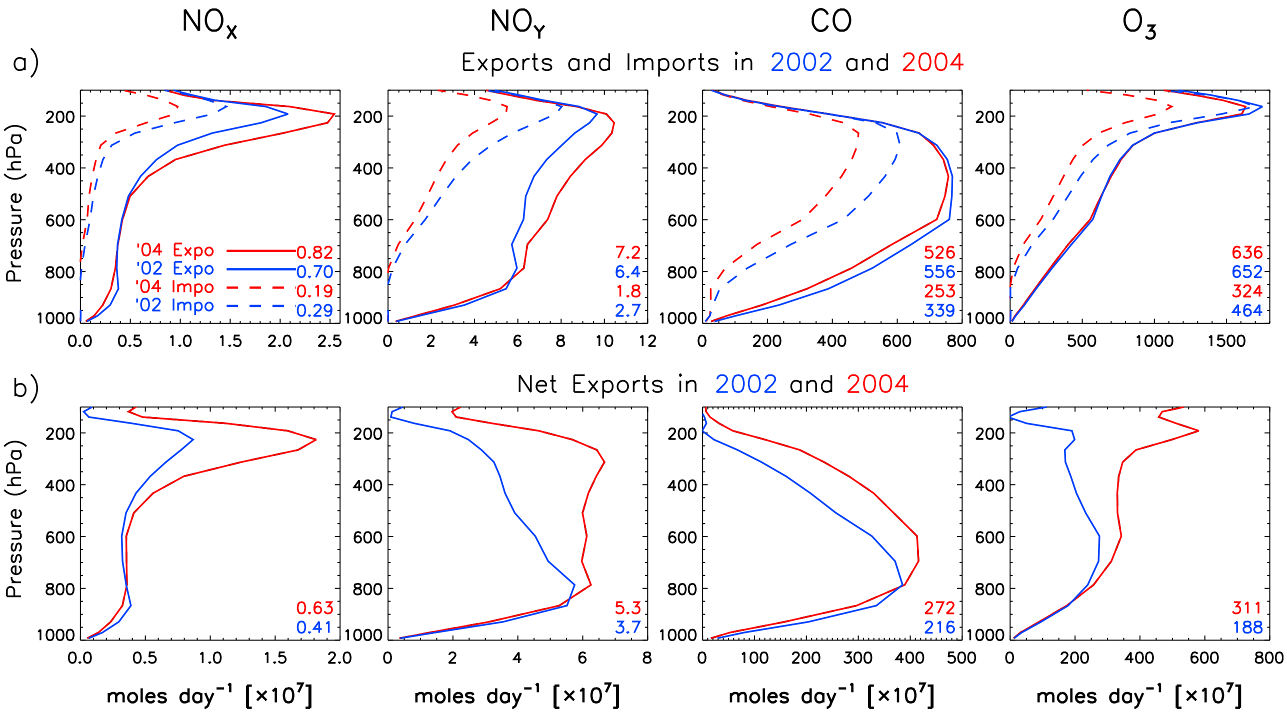

Figure 14. Early summer (1 June to 17 July) fluxes of NO_x, NO_y, CO, and O_3 across the western and eastern boundaries of North America (summed between 25°N and 60°N) at 130°W (imports) and 65°W (exports) from simulation L1 for (a) 2004 exports (red solid), 2004 imports (red dashed), 2002 exports (blue solid), and 2002 imports (blue dashed) and (b) 2004 (red) and 2002 (blue) net exports. Vertically averaged fluxes (from the surface to 100 hPa) are listed in the bottom right corner of each plot. Fluxes across the northern and southern boundaries of NA are small and are not shown.

[54] We see the largest impact from lightning after the onset of the North American Monsoon (Figure 12e), which is reinforced by the upper level anticyclone centered over Mexico [*Cooper et al.*, 2009, Figure 2]. Although the RF_{LNOx} is greater in late than early summer in the immediate convective outflow over Mexico and the Gulf of Mexico, it is slightly less over Europe and northern Africa in late summer 2004. This indicates that the convective outflow recirculated in this upper level anticyclone centered over Mexico in late summer, allowing more O_3 production over the southern United States and the Gulf of Mexico, as discussed by *Li et al.* [2005], prior to transport across the Atlantic.

[55] The spatial pattern of summer-to-summer changes in RF (Figures 12c and 12f) is similar to that of summer-to-summer changes in the tropospheric O_3 column. RF_{LNOx} was nearly a factor of 2 larger in early summer 2004 than 2002 in the NA outflow region. The impact of power plant reductions on RF (diagnosed from the sensitivity simulation with 2002 NO_x emissions and 2004 meteorology and lightning) was much smaller than that seen in Figures 12c and 12f. The RF values over the North Atlantic decreased by <0.01 W m^{-2} because of emissions reductions (not shown).

[56] In early summer 2004, mean normalized RF per unit of added O_3 column, over the areas with enhancements exceeding 5 DU, is 0.027 W m^{-2} DU^{-1} due to NA anthropogenic enhancements and 0.047 W m^{-2} DU^{-1} due to NA lightning enhancements (average from L1 and L2 simulations). For comparison with previous studies we use *Gauss et al.* [2003], who used 11 different climate models to estimate the (longwave, clear-sky) normalized instantaneous radiative forcing. They gave a range of 0.042–0.052 W m^{-2} DU^{-1} for the global annual averages of normalized radiative forcing due to changes in tropospheric O_3 between 2000 and 2100. The annual average of normalized radiative forcing due to increasing anthropogenic emissions over the next century is predicted to be greater than that due to present-day NA anthropogenic emissions and is comparable with RF due to NA lightning NO emissions. Noteworthy is also *Worden et al.* [2008], who, using TES (Tropospheric Emission Spectrometer) measurements for cloud-free ocean conditions, obtained an estimate of 0.055 W m^{-2} DU^{-1} (annual mean from 45°S to 45°N) for the sensitivity to UT (500–200 hPa) O_3, thus providing an important observational constraint for both natural and anthropogenic O_3.

3.4. Import and Export Fluxes

[57] We calculate the fluxes of NO_x, NO_y, CO and O_3 across the western and eastern NA boundaries for early summer. These fluxes are summed along the longitudes 130°W (imports) and 65°W (exports) in the region between 25°N and 60°N. We estimate NO_y as the sum of the following oxidation products: NO_x, NO_3, N_2O_5, HONO, HNO_3, HO_2NO_2, PAN and MPAN (methylperoxyacetyl nitrate).

[58] Figure 14a shows vertical profiles of NO_x, NO_y, CO and O_3 fluxes averaged over early summer 2002 and

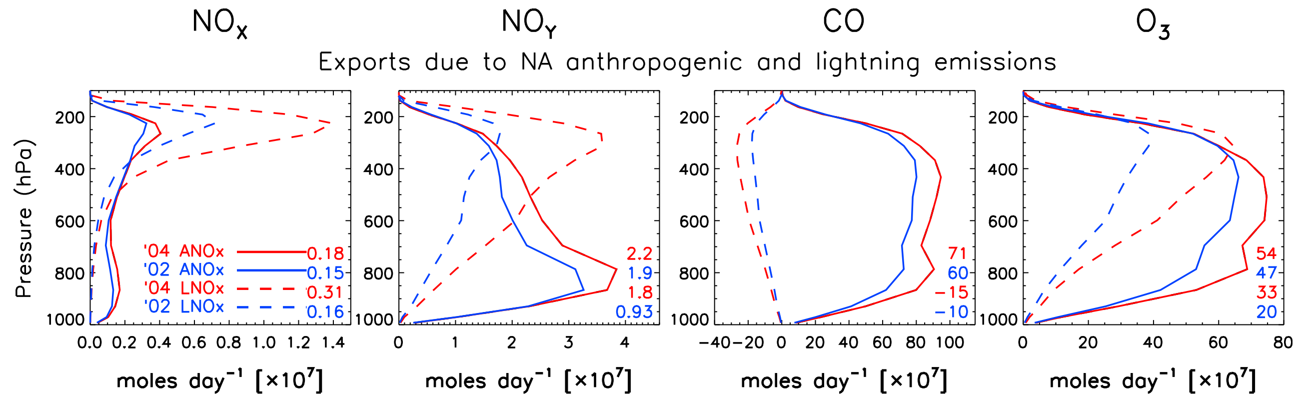

Figure 15. Same as Figure 14 except that exports due to NA anthropogenic (solid) and lightning (dashed) emissions are shown as diagnosed by sensitivity simulations with respective sources turned off (Table 1). Exports in 2004 are red and exports in 2002 are blue.

2004; 2002 imports exceeded 2004 imports for each species. Since we held the Asian anthropogenic emissions constant for both years, the decreased imports (by ~30%) in 2004 compared to 2002 were mainly due to the weaker jet stream over the Pacific. For example the peak zonal winds at 225 hPa were 36 m s^{-1} and 32 m s^{-1} in 2002 and 2004, respectively, although it should be noted that the peak zonal winds do not represent the integrated effect of the entire wind field over the Pacific. The reduced imports seen in 2004 compared to 2002 motivate us to calculate the difference between exports and imports, hereafter referred to as the net exports. This method allows us to estimate the efficiency of photochemistry over North America by removing the differences in what is imported from what is photochemically produced or emitted over North America. O_3 soundings at the west coast site of Trinidad Head, California, indicate that O_3 imports are well simulated (Figure S4 in the auxiliary material shows a good agreement of ozonesonde-measured O_3 profiles with model-calculated O_3 profiles).

[59] NO_x and NO_y fluxes in this study are similar to those from *Choi et al.* [2008], in which the authors estimated imports and exports from North America in spring 2000 using the regional chemistry transport model REAM. They reported that in May 2000, the NO_x exports peak at 4 × 10^7 mol d^{-1}. In our study, NO_x exports peak at 2.1–2.5 × 10^7 mol d^{-1} (in early summer 2002 or 2004, Figure 14a). This lower peak could be because of faster photochemical oxidation and slower wind speeds during summer. In addition, CEMS data show that NO_x emissions from the United States were greater in 2000 than 2002 or 2004. North America is a net source of pollution in summer (exports greater than imports throughout the troposphere); this is partially due to stronger westerlies over the western Atlantic than over the eastern Pacific (Figure 3). The total O_3 exports (summed from the surface to 100 hPa) were factors of 2 and 1.4 larger than the total O_3 imports in early summer 2004 and 2002, respectively, whereas in May 2000, the export-to-import ratio was close to 1 [*Choi et al.*, 2008].

[60] Figure 14b indicates that the net exports were larger in early summer 2004 than in 2002. The areas to the left of the net export curves are proportional to the total mass exported from North America. CO was exported at higher altitudes in 2004 (Figure 14b) which along with enhanced westerlies (Figure 3) led to greater net export than in 2002. In order to determine if differences in biomass burning between 2002 and 2004 had a substantial impact on net exports, we reran the UMD-CTM for early summer 2004 using 2002 biomass burning emissions. Net CO exports increased by 9%; the impact on other species was smaller. Because of stronger westerlies, enhanced lightning NO emissions over North America in summer 2004 than in 2002 and a possible increase in O_3 imported from stratosphere in 2004 (consistent with *Thompson et al.* [2007a]), net O_3 exports were greatly enhanced in the UT compared to 2002.

[61] Despite reduced ANO_x emissions due to the NO_x SIP Call and cooler temperatures (Figure 3b) in 2004 relative to 2002, simulations with the UMD-CTM show greater anthropogenic exports in 2004 than in 2002 (Figure 15a). This was likely due to an efficient transport mechanism from North America: enhanced convective lofting over polluted areas (Figure 2b) and stronger westerlies (Figures 3b and 3d). Similarly, the exports of NO_x, NO_y and O_3 due to lightning NO emissions in 2004 greatly exceeded those seen in 2002 (Figure 15).

[62] Because of lofted pollution from the BL, ANO_x exports peaked in the UT in both years. NO_y exports due to anthropogenic emissions peaked in the LT. Increased lightning activity and stronger UT westerlies over the CONUS in early summer 2004 resulted in a factor of 2 greater LNO_x exports than in 2002. In early summer 2004, anthropogenic emissions explain about 28% and 41% of the net NO_x and NO_y column exports, respectively, while lightning explains about 49%–67% and 34%–49% (the lower estimates corresponding to production of 240 NO mol $flash^{-1}$ and the upper estimates corresponding to production of 480 NO mol $flash^{-1}$). The remaining 5%–23% of the net NO_x, and 10%–25% of the net NO_y, is from other NO_x sources (biomass burning emissions and soil release) and the contribution from the north and south into North America. O_3 exports due to anthropogenic emissions were a factor of 1.6 larger than those due to LNO_x emissions in 2004 (54 × 10^7 O_3 mol d^{-1} compared to 33 × 10^7 O_3 mol d^{-1}). However, in a sensitivity simulation with doubled LNO_x source, this ratio decreases to 1.2 (O_3 exports due to lightning increased by 33% to 44 × 10^7 O_3 mol d^{-1}). This is consistent with the earlier result

where doubled LNO_x source over North America increased the lightning enhancements by ~33% in downwind regions compared to lightning enhancements from LNO_x source in the standard simulation. In 2002, anthropogenic emissions contributed twice as much to the net O_3 export as lightning.

4. Summary

[63] This study illustrates the importance of interannual variations in meteorology and associated lightning for the variability of long-range transport and continental outflow. We conducted several simulations of summers 2002 and 2004 with the UMD-CTM driven by meteorological fields from the GEOS-4 CERES reanalysis. Summer 2004 had reduced power plant NO_x emissions in the Ohio River Valley (resulting from the NO_x SIP Call), more lightning, relatively cool temperatures and frequent synoptic disturbances over the contiguous United States compared to summer 2002. We used 2004 as a reference year to evaluate the UMD-CTM due to the valuable measurements that were obtained over the eastern United States and western North Atlantic during the INTEX-A science mission. Summer 2004 revealed great decreases in observed O_3 concentrations over the northeastern United States, especially downwind of the Ohio River Valley, the region with a high number of power plants that had implemented NO_x controls. The satellite observations from SCIAMACHY clearly detected NO_2 column decreases in this region.

[64] We also conducted several lightning sensitivity simulations. We assumed the lightning flashes are proportional to the square of convective mass fluxes and constrained to match the observations from OTD/LIS and from the NLDN. We found an inconsistency between LIS- and NLDN-based total flash rates over the CONUS south of 35°N: more lightning was observed by the NLDN network (after adjustment by the IC/CG ratios) than by the LIS sensor. For agreement between these two data sets, the summertime IC/CG ratios over this region would have to be decreased by a factor of 2.

[65] Like other global and regional CTMs, O_3 in the UMD-CTM is overestimated by 15–25 ppbv at the surface and by up to 12 ppbv in the upper troposphere (500–200 hPa) compared to aircraft and ozonesonde measurements. We found that the simulation with doubled lightning NO production (480 mol flash^{-1}) agrees best with observed NO_x; however, it increases the upper tropospheric high bias for O_3 by ~3 ppbv. Because of these mixed results, we complement the results from the standard simulation with the results from the simulation with doubled lightning NO production per flash. In the Ohio River Valley, the UMD-CTM showed similar reductions (19%) of tropospheric NO_2 column as observed by SCIAMACHY (22%) between August 2002 and 2004, consistent with the emission reductions due to the NO_x SIP Call. Simulations with the UMD-CTM showed reduced O_3 concentrations at the surface between the summers 2002 and 2004; however, these reductions were 50% less than those seen in AQS observations.

[66] Lightning over the United States greatly enhances the North American outflow of O_3. In early summer 2004, North American anthropogenic emissions produced the greatest O_3 enhancements near the surface (up to 35 ppbv over the eastern United States), whereas lightning had the greatest impact in the upper troposphere (up to 16 ppbv for the standard and 20 ppbv for doubled LNO_x source, over the Gulf Coast and the western North Atlantic). After the onset of the North American Monsoon, the impact of lightning was even greater (up to 18 ppbv for the standard and 22 ppbv for doubled LNO_x source). RF (defined as net downward radiative flux at the tropopause for clear-sky conditions) of 0.15–0.30 W m^{-2} due to O_3 produced from anthropogenic emissions was seen in the continental outflow across the North Atlantic, extending to Europe and northern Africa, while RF due to O_3 produced from lightning NO emissions was 0.20–0.40 W m^{-2} (0.25–0.50 W m^{-2} for doubled LNO_x source) over the same areas in early and late summer 2004. Lightning flash rates in early summer 2004 were 50% higher than in 2002 over the contiguous United States. RF due to lightning was nearly a factor of 2 larger in early summer 2004 than 2002 in the North American outflow region. The normalized RF per unit of added O_3 column was 0.027 W m^{-2} DU^{-1} for anthropogenic enhancements and 0.047 W m^{-2} DU^{-1} for lightning enhancements. This is because of stronger radiative forcing efficiency of an upper tropospheric perturbation.

[67] Sensitivity simulation with reduced emissions due to the NO_x SIP Call showed that the impact of emission reductions (between 2002 and 2004) on O_3 column over the North Atlantic (30°N–50°N) was a factor of 7 smaller than the impact of changes in lightning and meteorology in early summer. Late summer changes in lightning had much smaller impact on O_3 columns.

[68] Large differences between the two summers in horizontal winds and convection greatly modulated the changes in O_3 concentrations. Simulations with the UMD-CTM show that despite reduced emissions due to the NO_x SIP Call and cooler temperatures in 2004 relative to 2002, more O_3 was exported from North America in 2004 due to anthropogenic emissions than in 2002 because of enhanced lofting of polluted air from the boundary layer (in early summer) followed by stronger westerly winds over the main NO_x source region in the eastern United States. O_3 exports across the eastern NA boundary due to anthropogenic emissions were factor of 1.6 larger than those due to lightning in 2004. However, the simulation with doubled lightning source reduces this ratio to only 1.2 indicating nonlinearity. Doubling the North American lightning NO source increased downwind O_3 enhancements due to lightning NO emissions by one third.

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References

Allen, D. J., R. B. Rood, A. M. Thompson, and R. D. Hudson (1996), Three-dimensional radon 222 calculations using assimilated meteorological data and a convective mixing algorithm, *J. Geophys. Res.*, *101*(D3), 6871–6881, doi:10.1029/95JD03408.

Allen, D. J., K. E. Pickering, J. Rodriguez, B. N. Duncan, S. E. Strahan, and M. Damon (2010), Impact of lightning NO emissions on North American photochemistry as determined using the Global Modeling Initiative (GMI) model, *J. Geophys. Res.*, *115*, D22301, doi:10.1029/2010JD014062.

Andreae, M. O., and P. Merlet (2001), Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cycles*, *15*(4), 955–966, doi:10.1029/2000GB001382.

Bertram, T. H., et al. (2007), Direct measurements of the convective recycling of the upper troposphere, *Science*, *315*(5813), 816–820, doi:10.1126/science.1134548.

Biagi, C. J., K. L. Cummins, K. E. Kehoe, and E. P. Krider (2007), National Lightning Detection Network (NLDN) performance in southern Arizona, Texas, and Oklahoma in 2003–2004, *J. Geophys. Res.*, *112*, D05208, doi:10.1029/2006JD007341.

Blitz, M. A., D. E. Heard, M. J. Pilling, S. R. Arnold, and M. P. Chipperfield (2004), Pressure and temperature-dependent quantum yields for the photodissociation of acetone between 279 and 327.5 nm, *Geophys. Res. Lett.*, *31*, L06111, doi:10.1029/2003GL018793. (Correction, *Geophys. Res. Lett.*, *31*, L09104, doi:10.1029/2004GL020182, 2004.)

Bloom, S., et al. (2005), Documentation and validation of the Goddard Earth Observing System (GEOS) data assimilation system—Version 4, *NASA TM-2005-104606*, 26 pp.

Boccippio, D. J., K. L. Cummins, H. J. Christian, and S. J. Goodman (2001), Combined satellite- and surface-based estimation of the intracloud-cloud-to-ground lightning ratio over the continental United States, *Mon. Weather Rev.*, *129*(1), 108–122, doi:10.1175/1520-0493(2001)129<0108:CSASBE>2.0.CO;2.

Boccippio, D. J., W. J. Koshak, and R. J. Blakeslee (2002), Performance assessment of the optical transient detector and lightning Imaging sensor. Part I: Predicted diurnal variability, *J. Atmos. Oceanic Technol.*, *19*(9), 1318–1332, doi:10.1175/1520-0426(2002)019<1318:PAOTOT>2.0.CO;2.

Bousserez, N., et al. (2007), Evaluation of the MOCAGE chemistry transport model during the ICARTT/ITOP experiment, *J. Geophys. Res.*, *112*, D10S42, doi:10.1029/2006JD007595.

Büker, M. L., M. H. Hitchman, G. J. Tripoli, R. B. Pierce, E. V. Browell, and J. A. Al-Saadi (2008), Long-range convective ozone transport during INTEX, *J. Geophys. Res.*, *113*, D14S90, doi:10.1029/2007JD009345.

Choi, Y., Y. Wang, T. Zeng, D. Cunnold, E.-S. Yang, R. Martin, K. Chance, V. Thouret, and E. Edgerton (2008), Springtime transitions of NO_2, CO, and O_3 over North America: Model evaluation and analysis, *J. Geophys. Res.*, *113*, D20311, doi:10.1029/2007JD009632.

Choi, Y., J. Kim, A. Eldering, G. Osterman, Y. L. Yung, Y. Gu, and K. N. Liou (2009), Lightning and anthropogenic NO_x sources over the United States and the western North Atlantic Ocean: Impact on OLR and radiative effects, *Geophys. Res. Lett.*, *36*, L17806, doi:10.1029/2009GL039381.

Chou, M.-D., and M. Suarez (1994), An efficient thermal infrared radiation parameterization for use in general circulation models, *NASA Tech. Memo.*, *TM-104606*, 85 pp.

Chou, M.-D., W. L. Ridgway, and M. M. H. Yan (1995), Parameterizations for water vapor IR radiative transfer in both the middle and lower atmospheres, *J. Atmos. Sci.*, *52*(8), 1159–1167, doi:10.1175/1520-0469(1995)052<1159:PFWVIR>2.0.CO;2.

Christian, H. J., et al. (2003), Global frequency and distribution of lightning as observed from space by the Optical Transient Detector, *J. Geophys. Res.*, *108*(D1), 4005, doi:10.1029/2002JD002347.

Cooper, O. R., et al. (2006), Large upper tropospheric ozone enhancements above midlatitude North America during summer: In situ evidence from the IONS and MOZAIC ozone measurement network, *J. Geophys. Res.*, *111*, D24S05, doi:10.1029/2006JD007306.

Cooper, O. R., et al. (2009), Summertime buildup and decay of lightning NO_x and aged thunderstorm outflow above North America, *J. Geophys. Res.*, *114*, D01101, doi:10.1029/2008JD010293.

Cummins, K. L., M. J. Murphy, E. A. Bardo, W. L. Hiscox, R. B. Pyle, and A. E. Pifer (1998), A combined TOA/MDF technology upgrade of the U.S. National Lightning Detection Network, *J. Geophys. Res.*, *103*(D8), 9035–9044, doi:10.1029/98JD00153.

Cummins, K. L., et al. (2006), The U.S. National Lightning Detection Network: Post-upgrade status, paper presented at 2nd Conference on Meteorological Applications of Lightning Data, Am. Meteorol. Soc., Atlanta, Ga.

DeCaria, A. J., K. E. Pickering, G. L. Stenchikov, J. R. Scala, J. L. Stith, J. E. Dye, B. A. Ridley, and P. Laroche (2000), A cloud-scale model study of lightning-generated NO_x in an individual thunderstorm during STERAO-A, *J. Geophys. Res.*, *105*(D9), 11,601–11,616, doi:10.1029/2000JD900033.

DeCaria, A. J., K. E. Pickering, G. L. Stenchikov, and L. E. Ott (2005), 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*, D14303, doi:10.1029/2004JD005556.

Dickerson, R. R., et al. (1987), Thunderstorms: An important mechanism in the transport of air pollutants, *Science*, *235*(4787), 460–4654, doi:10.1126/science.235.4787.460.

Douglass, A. R., R. S. Stolarski, S. E. Strahan, and P. S. Connell (2004), Radicals and reservoirs in the GMI chemistry and transport model: Comparison to measurements, *J. Geophys. Res.*, *109*, D16302, doi:10.1029/2004JD004632.

Environmental Protection Agency (2005), Evaluating ozone control programs in the eastern United States: Focus on the NO_x budget trading program, 2004, 31 pp., Washington, D. C. (Available at http://www.epa.gov/airtrends/2005/ozonenbp)

Fang, Y., A. M. Fiore, L. W. Horowitz, H. Levy II, Y. Hu, and A. G. Russell (2010), Sensitivity of the NO_y budget over the United States to anthropogenic and lightning NO_x in summer, *J. Geophys. Res.*, *115*, D18312, doi:10.1029/2010JD014079.

Fehsenfeld, F. C., et al. (2006), International Consortium for Atmospheric Research on Transport and Transformation (ICARTT): North America to Europe—Overview of the 2004 summer field study, *J. Geophys. Res.*, *111*, D23S01, doi:10.1029/2006JD007829.

Friedl, M. A., et al. (2002), Global land cover mapping from MODIS: Algorithms and early results, *Remote Sens. Environ.*, *83*, 287–302, doi:10.1016/S0034-4257(02)00078-0.

Frost, G. J., et al. (2006), Effects of changing power plant NO_x emissions on ozone in the eastern United States: Proof of concept, *J. Geophys. Res.*, *111*, D12306, doi:10.1029/2005JD006354.

Fuelberg, H. E., M. J. Porter, C. M. Kiley, J. J. Halland, and D. Morse (2007), Meteorological conditions and anomalies during the Intercontinental Chemical Transport Experiment–North America, *J. Geophys. Res.*, *112*, D12S06, doi:10.1029/2006JD007734.

Gao, X., J. Li, and S. Sorooshian (2007), Modeling intraseasonal features of 2004 North American monsoon precipitation, *J. Clim.*, *20*(9), 1882–1896, doi:10.1175/JCLI4100.1.

Gauss, M., et al. (2003), Radiative forcing in the 21st century due to ozone changes in the troposphere and the lower stratosphere, *J. Geophys. Res.*, *108*(D9), 4292, doi:10.1029/2002JD002624.

Giglio, L., G. R. van der Werf, J. T. Randerson, G. J. Collatz, and P. Kasibhatla (2006), Global estimation of burned area using MODIS active fire observations, *Atmos. Chem. Phys.*, *6*, 957–974, doi:10.5194/acp-6-957-2006.

Gilliland, A. B., C. Hogrefe, R. W. Pinder, J. M. Godowitch, K. L. Foley, and S. T. Rao (2008), Dynamic evaluation of regional air quality models: Assessing changes in O_3 stemming from changes in emissions and meteorology, *Atmos. Environ.*, *42*(20), 5110–5123, doi:10.1016/j.atmosenv.2008.02.018.

Godowitch, J. M., A. B. Gilliland, R. R. Draxler, and S. T. Rao (2008), Modeling assessment of point source NO_x emission reductions on ozone air quality in the eastern United States, *Atmos. Environ.*, *42*, 87–100, doi:10.1016/j.atmosenv.2007.09.032.

Guenther, A., T. Karl, P. Harley, C. Wiedinmyer, P. I. Palmer, and C. Geron (2006), Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, *6*, 3181–3210, doi:10.5194/acp-6-3181-2006.

Hack, J. J. (1994), Parameterization of moist convection in the National Center for Atmospheric Research community climate model (CCM2), *J. Geophys. Res.*, *99*(D3), 5551–5568, doi:10.1029/93JD03478.

Hansen, M. C., R. S. Defries, J. R. G. Townshend, and R. Sohlberg (2000), Global land cover classification at 1km spatial resolution using a classification tree approach, *Int. J. Remote Sens.*, *21*, 1331–1364, doi:10.1080/014311600210209.

Heald, C. L., D. J. Jacob, P. I. Palmer, M. J. Evans, G. W. Sachse, H. B. Singh, and D. R. Blake (2003), Biomass burning emission inventory with daily resolution: Application to aircraft observations of Asian outflow, *J. Geophys. Res.*, *108*(D21), 8811, doi:10.1029/2002JD003082.

Henderson, B., et al. (2010), Evaluation of simulated photochemical partitioning of oxidized nitrogen in the upper troposphere, *Atmos. Chem. Phys. Discuss.*, *10*, 20,125–20,165, doi:10.5194/acpd-10-20125-2010.

Hogrefe, C., B. Lynn, K. Civerolo, J.-Y. Ku, J. Rosenthal, C. Rosenzweig, R. Goldberg, S. Gaffin, K. Knowlton, and P. L. Kinney (2004), Simulating changes in regional air pollution over the eastern United States due to changes in global and regional climate and emissions, *J. Geophys. Res.*, *109*, D22301, doi:10.1029/2004JD004690.

Hudman, R. C., et al. (2007), Surface and lightning sources of nitrogen oxides over the United States: Magnitudes, chemical evolution, and outflow, *J. Geophys. Res.*, *112*, D12S05, doi:10.1029/2006JD007912.

Hudman, R. C., L. T. Murray, D. J. Jacob, S. Turquety, S. Wu, D. B. Millet, M. Avery, A. H. Goldstein, and J. Holloway (2009), North American influence on tropospheric ozone and the effects of recent emission reductions: Constraints from ICARTT observations, *J. Geophys. Res.*, *114*, D07302, doi:10.1029/2008JD010126.

Hyer, E. J., E. S. Kasischke, and D. J. Allen (2007), Effects of source temporal resolution on transport simulations of boreal fire emissions, *J. Geophys. Res.*, *112*, D01302, doi:10.1029/2006JD007234.

Intergovernmental Panel on Climate Change (IPCC) (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., 996 pp., Cambridge Univ. Press, Cambridge, U. K.

Jourdain, L., S. S. Kulawik, H. M. Worden, K. E. Pickering, J. Worden, and A. M. Thompson (2010), Lightning NO_x emissions over the USA investigated using TES ozone observations and the GEOS-Chem model, *Atmos. Chem. Phys.*, *10*, 107–119, doi:10.5194/acp-10-107-2010.

Jet Propulsion Laboratory (2006), Chemical kinetics and photochemical data for use in atmospheric studies, *JPL Publ., 06-2*, Pasadena, Calif.

Justice, C. O., et al. (2002), The MODIS fire products, *Remote Sens. Environ.*, *83*, 244–262, doi:10.1016/S0034-4257(02)00076-7.

Kasischke, E. S., E. J. Hyer, P. C. Novelli, L. P. Bruhwiler, N. H. F. French, A. I. Sukhinin, J. H. Hewson, and B. J. Stocks (2005), Influences of boreal fire emissions on Northern Hemisphere atmospheric carbon and carbon monoxide, *Global Biogeochem. Cycles*, *19*, GB1012, doi:10.1029/2004GB002300.

Kiley, C. M., and H. E. Fuelberg (2006), An examination of summertime cyclone transport processes during Intercontinental Chemical Transport Experiment (INTEX-A), *J. Geophys. Res.*, *111*, D24S06, doi:10.1029/2006JD007115.

Kim, S. W., A. Heckel, S. A. McKeen, G. J. Frost, E.-Y. Hsie, M. K. Trainer, A. Richter, J. P. Burrows, S. E. Peckham, and G. A. Grell (2006), Satellite-observed U.S. power plant NO_x emission reductions and their impact on air quality, *Geophys. Res. Lett.*, *33*, L22812, doi:10.1029/2006GL027749.

Labrador, L. J., R. von Kuhlmann, and M. G. Lawrence (2005), 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, doi:10.5194/acp-5-1815-2005.

Lacis, A. A., D. J. Wuebbles, and J. A. Logan (1990), Radiative forcing of climate by changes in the vertical distribution of ozone, *J. Geophys. Res.*, *95*(D7), 9971–9981, doi:10.1029/JD095iD07p09971.

Lelieveld, J., and P. J. Crutzen (1994), Role of deep cloud convection in the ozone budget of the troposphere, *Science*, *264*(5166), 1759–1761, doi:10.1126/science.264.5166.1759.

Li, J., X. Gao, R. A. Maddox, and S. Sorooshian (2004), Model study of evolution and diurnal variations of rainfall in the north American monsoon during June and July 2002, *Mon. Weather Rev.*, *132*(12), 2895–2915, doi:10.1175/MWR2832.1.

Li, Q. B., et al. (2002), Transatlantic transport of pollution and its effects on surface ozone in Europe and North America, *J. Geophys. Res.*, *107*(D13), 4166, doi:10.1029/2001JD001422.

Li, Q., D. J. Jacob, R. Park, Y. Wang, C. L. Heald, R. Hudman, R. M. Yantosca, R. V. Martin, and M. Evans (2005), North American pollution outflow and the trapping of convectively lifted pollution by upper-level anticyclone, *J. Geophys. Res.*, *110*, D10301, doi:10.1029/2004JD005039.

Liu, H. Y., D. J. Jacob, I. Bey, and R. M. Yantosca (2001), Constraints from ^{210}Pb and ^{7}Be on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields, *J. Geophys. Res.*, *106*(D11), 12,109–12,128, doi:10.1029/2000JD900839.

Mach, D. M., H. J. Christian, R. J. Blakeslee, D. J. Boccippio, S. J. Goodman, and W. L. Boeck (2007), Performance assessment of the Optical Transient Detector and Lightning Imaging Sensor, *J. Geophys. Res.*, *112*, D09210, doi:10.1029/2006JD007787.

McLinden, C. A., S. C. Olsen, B. Hannegan, O. Wild, M. J. Prather, and J. Sundet (2000), Stratospheric ozone in 3-D models: A simple chemistry and the cross-tropopause flux, *J. Geophys. Res.*, *105*(D11), 14,653–14,665, doi:10.1029/2000JD900124.

Nolte, C. G., A. B. Gilliland, C. Hogrefe, and L. J. Mickley (2008), Linking global to regional models to assess future climate impacts on surface ozone levels in the United States, *J. Geophys. Res.*, *113*, D14307, doi:10.1029/2007JD008497.

Olivier, J. G. J., and J. J. M. Berdowski (2001), Global emissions sources and sinks, in *The Climate System*, edited by J. Berdowski, R. Guicherit, and B. J. Heij, pp. 33–78, A. A. Balkema, Lisse, Netherlands.

Olivier, J. G. J., J. A. van Aardenne, F. Dentener, L. Ganzeveld, and J. A. H. W. Peters (2005), Recent trends in global greenhouse gas emissions: Regional trends and spatial distribution of key sources, in *Non-CO_2 Greenhouse Gases (NCGG-4)*, edited by A. van Amstel, pp. 325–330, Millpress, Rotterdam, Netherlands. (Available at http://www.mnp.nl/edgar/Images/Olivier2005-FT2000-NCGG4-Utrecht_tcm32-22124.pdf)

Orville, R. E., and G. R. Huffines (2001), Cloud-to-ground lightning in the United States: NLDN results in the first decade, 1989–98, *Mon. Weather Rev.*, *129*(5), 1179–1193, doi:10.1175/1520-0493(2001)129<1179:CTGLIT>2.0.CO;2.

Ott, L. E., K. E. Pickering, G. L. Stenchikov, D. J. Allen, A. J. DeCaria, B. Ridley, R.-F. Lin, S. Lang, and W.-K. Tao (2010), Production of lightning NO and its vertical distribution calculated from three-dimensional cloud-scale chemical transport model simulations, *J. Geophys. Res.*, *115*, D04301, doi:10.1029/2009JD011880.

Pacifico, F., S. P. Harrison, C. D. Jones, and S. Sitch (2009), Isoprene emissions and climate, *Atmos. Environ.*, *43*(39), 6121–6135, doi:10.1016/j.atmosenv.2009.09.002.

Park, R. J., G. L. Stenchikov, K. E. Pickering, R. R. Dickerson, D. J. Allen, and S. Kondragunta (2001), Regional air pollution and its radiative forcing: Studies with a single-column chemical and radiation transport model, *J. Geophys. Res.*, *106*(D22), 28,751–28,770, doi:10.1029/2001JD001182.

Park, R. J., K. E. Pickering, D. J. Allen, G. L. Stenchikov, and M. S. Fox-Rabinovitz (2004a), Global simulation of tropospheric ozone using the University of Maryland Chemical Transport Model (UMD-CTM): 1. Model description and evaluation, *J. Geophys. Res.*, *109*, D09301, doi:10.1029/2003JD004266.

Park, R. J., K. E. Pickering, D. J. Allen, G. L. Stenchikov, and M. S. Fox-Rabinovitz (2004b), Global simulation of tropospheric ozone using the University of Maryland Chemical Transport Model (UMD-CTM): 2. Regional transport and chemistry over the central United States using a stretched grid, *J. Geophys. Res.*, *109*, D09303, doi:10.1029/2003JD004269.

Pickering, K. E., A. M. Thompson, J. R. Scala, W. K. Tao, R. R. Dickerson, and J. Simpson (1992), Free tropospheric ozone production following entrainment of urban plumes into deep convection, *J. Geophys. Res.*, *97*(16), 17,985–18,000.

Pickering, K. E., A. M. Thompson, W. K. Tao, R. B. Rood, D. P. McNamara, and A. M. Molod (1995), Vertical transport by convective clouds: Comparisons of three modeling approaches, *Geophys. Res. Lett.*, *22*(9), 1089–1092, doi:10.1029/95GL00889.

Pickering, K. E., Y. S. Wang, and W. K. Tao (1998), Vertical distributions of lightning NO_x for use in regional and global chemical transport models, *J. Geophys. Res.*, *103*(D23), 31,203–31,216, doi:10.1029/98JD02651.

Pierce, R. B., et al. (2007), Chemical data assimilation estimates of continental U.S. ozone and nitrogen budgets during the Intercontinental Chemical Transport Experiment–North America, *J. Geophys. Res.*, *112*, D12S21, doi:10.1029/2006JD007722.

Reidmiller, D. R., et al. (2009), The influence of foreign vs. North American emissions on surface ozone in the US, *Atmos. Chem. Phys.*, *9*(14), 5027–5042, doi:10.5194/acp-9-5027-2009.

Ren, X. R., et al. (2008), HO_x chemistry during INTEX-A 2004: Observation, model calculation, and comparison with previous studies, *J. Geophys. Res.*, *113*, D05310, doi:10.1029/2007JD009166.

Richter, A., J. P. Burrows, H. Nuss, C. Granier, and U. Niemeier (2005), Increase in tropospheric nitrogen dioxide over China observed from space, *Nature*, *437*(7055), 129–132, doi:10.1038/nature04092.

Ridley, B. A., J. G. Walega, J. E. Dye, and F. E. Grahek (1994), Distribution of NO, NO_x, NO_y, and O_3 to 12 km altitude during the summer monsoon season over New Mexico, *J. Geophys. Res.*, *99*(D12), 25,519–25,534, doi:10.1029/94JD02210.

Roy, B., et al. (2007), Refining fire emissions for air quality modeling with remotely sensed fire counts: A wildfire case study, *Atmos. Environ.*, *41*, 655–665, doi:10.1016/j.atmosenv.2006.08.037.

Schumann, U., and H. Huntrieser (2007), The global lightning-induced nitrogen oxides source, *Atmos. Chem. Phys.*, *7*(14), 3823–3907, doi:10.5194/acp-7-3823-2007.

Sillman, S., J. A. Logan, and S. C. Wofsy (1990), Regional-scale model for ozone in the United States with subgrid representation of urban and power plant plumes, *J. Geophys. Res.*, *95*(D5), 5731–5748, doi:10.1029/JD095iD05p05731.

Singh, H. B., W. H. Brune, J. H. Crawford, D. J. Jacob, and P. B. Russell (2006), Overview of the summer 2004 Intercontinental Chemical Transport Experiment– North America (INTEX-A), *J. Geophys. Res.*, *111*, D24S01, doi:10.1029/2006JD007905.

Singh, H. B., et al. (2007), Reactive nitrogen distribution and partitioning in the North American troposphere and lowermost stratosphere, *J. Geophys. Res.*, *112*, D12S04, doi:10.1029/2006JD007664.

Thompson, A. M., et al. (2007a), Intercontinental Chemical Transport Experiment Ozonesonde Network Study (IONS) 2004: 1. Summertime upper troposphere/lower stratosphere ozone over northeastern North America, *J. Geophys. Res.*, *112*, D12S12, doi:10.1029/2006JD007441.

Thompson, A. M., et al. (2007b), Intercontinental Chemical Transport Experiment Ozonesonde Network Study (IONS) 2004: 2. Tropospheric ozone budgets and variability over northeastern North America, *J. Geophys. Res.*, *112*, D12S13, doi:10.1029/2006JD007670.

van der A, R. J., D. H. M. U. Peters, H. Eskes, K. F. Boersma, M. Van Roozendael, I. De Smedt, and H. M. Kelder (2006), Detection of the trend and seasonal variation in tropospheric NO_2 over China, *J. Geophys. Res.*, *111*, D12317, doi:10.1029/2005JD006594.

van der Werf, G. R., J. T. Randerson, G. J. Collatz, and L. Giglio (2003), Carbon Emissions from fires in tropical and subtropical ecosystems, *Global Change Biol.*, *9*(4), 547–562, doi:10.1046/j.1365-2486.2003.00604.x.

van der Werf, G. R., J. T. Randerson, L. Giglio, G. J. Collatz, P. S. Kasibhatla, and A. F. Arellano Jr. (2006), Interannual variability in global biomass burning emissions from 1997 to 2004, *Atmos. Chem. Phys.*, *6*, 3423–3441, doi:10.5194/acp-6-3423-2006.

Worden, H. M., K. W. Bowman, J. R. Worden, A. Eldering, and R. Beer (2008), Satellite measurements of the clear-sky greenhouse effect from tropospheric ozone, *Nat. Geosci.*, doi:10.1038/ngeo182.

Wu, S. L., B. N. Duncan, D. J. Jacob, A. M. Fiore, and O. Wild (2009), Chemical nonlinearities in relating intercontinental ozone pollution to anthropogenic emissions, *Geophys. Res. Lett.*, *36*, L05806, doi:10.1029/2008GL036607.

Zhang, G. J., and N. A. McFarlane (1995), Sensitivity of climate simulations to the parameterization of cumulus convection in the Canadian Climate Centre general circulation model, *Atmos. Ocean*, *33*, 407–446.

Zhang, R., T. Xuexi, and D. W. Bond (2003), Impacts of anthropogenic and natural NO_x sources over the U.S. on tropospheric chemistry, *Proc. Natl. Acad. Sci. U. S. A.*, *100*(4), 1505–1509, doi:10.1073/pnas.252763799.

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