IMPACT OF LIGHTNING-NO EMISSIONS ON EASTERN UNITED STATES PHOTOCHEMISTRY DURING THE SUMMER OF 2004 AS DETERMINED USING THE CMAQ MODEL

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1. INTRODUCTION

Production of NO by lightning (LNO_x) is an important part of the summertime tropospheric NO_x budget over the United States, but it is also its most uncertain component. Global model simulations indicate that LNO_x increases summertime upper tropospheric NO_x concentrations over the eastern United States by 60-75%, upper tropospheric ozone amounts by 15-25% and surface ozone concentrations by several ppbv [*Zhang et al.*, 2003; *Cooper et al.*, 2006; *D. J. Allen et al.* (manuscript in preparation, 2009)]. A simulation with the Community Multiscale Air Quality Model (CMAQ) that did not include lightning-NO emissions greatly underestimated upper tropospheric NO_x concentrations measured during NASA's INTEX-A field campaign. The simulation also underestimated nitric acid deposition at NADP sites by a factor of two. These large biases made it difficult to constrain ground-level NO_x emissions using SCIAMACHY retrievals and CMAQ output. A lightning-NO parameterization has been developed that can be used with the CMAQ model. Lightning-NO emissions in this scheme are assumed to be proportional to convective precipitation rate and scaled so that monthly average flash rates in each grid box match National Lightning Detection Network (NLDN) observed flash rates after adjusting for climatological intracloud to cloud-to-ground (IC/CG) ratios. The contribution of lightning-NO emissions to eastern United States NO_x and ozone distributions during the summer of 2004 will be evaluated by comparing results of CMAQ simulations with and without lightning-NO emissions to measurements from the NASA INTEX-A field campaign and to satellite retrievals from the SCIAMACHY instrument on the ENVISAT satellite.

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2. DESCRIPTION OF CMAQ SIMULATION

CMAQ [*Byun and Schere*, 2006] was used to simulate the concentrations of NO_2 and ozone as well as other pollutants in a continental US, 36 km horizontal resolution domain with 24 vertical layers from the surface to 100 hPa. Meteorological fields were developed using the fifth generation mesoscale model (MM5) version 3.6.3 [*Grell et al*., 1995], and the emissions inputs were the result of the Sparse Matrix Operator Kernel Emissions (SMOKE) version 2.0 processing of the 2002 National Emissions Inventory (NEI) for use with the Carbon Bond 2005 (CB-05) gas-phase chemical mechanism. The emissions included data from point sources equipped with continuous emissions monitoring systems (CEMs) that measure SO_2 and NO_x emission rates and other parameters daily, mobile emissions processed by the Mobile6 model, and meteorologically adjusted biogenic emissions from Biogenic Emission Inventory System (BEIS) 3.13 all specific for the year 2004.

3. SPECIFICATION OF LIGHTNING-NO SOURCE IN CMAQ

CMAQ requires emissions as a function of time and space. The lightning NO source (LNO_x) was parameterized in terms of the flash frequency (LF), flash energy (E), and the NO production per unit energy (P). Symbolically,

$$LNO_x = k * LF * E * P, \quad (1)$$

where k is a conversion factor equal to the molecular weight of nitrogen (N) divided by Avogadro's number. Implicit in this equation is the assumption that IC flashes are as energetic as CG flashes. The flash energy associated with CG and IC flashes has been the subject of much recent research. Recent field studies including STERAO [*DeCaria et al*., 2005], CRYSTAL-FACE [*Ott et al*., 2007]; and EULINOX [*Fehr et al.*, 2004] have found that IC flashes are approximately as energetic as CG flashes and that both CG and IC midlatitude flashes produce approximately 500 moles of N per flash on average.

In these simulations, we assume all flashes produce 500 moles of N. The flash frequency for each grid box is obtained by multiplying global (G) and local ($\alpha_{i,j}$) scaling factors by the convective precipitation rate minus a threshold. Symbolically,

$$LF(i,j) = G * \alpha(i,j) * (precon(i,j) – threshold), \qquad (2)$$

where *i* and *j* are indices of individual CMAQ grid boxes, *precon* is the convective precipitation rate from the MM5*, threshold* is the value of *precon* below which the flash rate is assumed to equal zero, *G* is a scaling factor chosen so that the domain-averaged MM5 flash rate equals a specified value, *α* is a local adjustment factor calculated after *G* and chosen so that the monthly average model-calculated flash rate for each grid box equals a specified value. For these simulations, G (α) is chosen so that domain-averaged (local) monthly average CMAQ flash rates matches flash rates obtained by multiplying monthly average CG flash rates from the NLDN [*Cummins et al*., 1998] by Z + 1, where Z is the monthly gridded climatological IC/CG ratio from *Boccippio et al.* [2001]. Figure 1 shows hourly CMAQ-calculated and NLDN-based total flash rates over the eastern US during August 2004. Both diurnal and day-to-day fluctuations in flash rates are reasonably well captured.

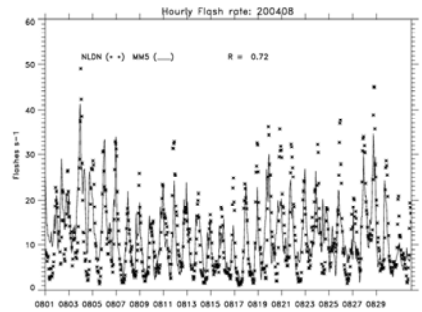

Figure 1. Hourly MM5-calculated total flash rates (solid lines) for the eastern US (110°-70° W, 25°-45° N) are compared to hourly NLDN-based total flash rates (asterisks) during August 2004.

LNO_x emissions are distributed in all model layers from the surface to the layer containing the convective cloud top. The mean April to September 2003-2005 vertical distribution of VHF sources (See Figure 2) from the Northern Alabama Lightning Mapping Array [*Koshak et al*., 2004] is used along with a direct proportionality to pressure to determine the percent of emissions apportioned to each layer.

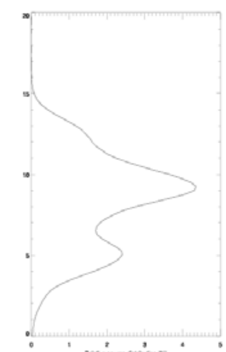

Figure 2. Mean vertical distribution of VHF sources from the Northern Alabama LMA for April to September 2003-2005 (courtesy of D. Buechler, NASA-MSFC).

3. COMPARISON WITH INTEX-A MEASUREMENTS

The INTEX-A field campaign was conducted from July 1 to August 15, 2004 over North America and the western Atlantic [*Singh et al*., 2006]. Its goals included source attribution. *Singh et al.* [2007] analyzed reactive nitrogen measurements during INTEX-A. They found unexpectedly large amounts of NO_x in the upper troposphere and suggested that lightning-NO emissions are a "far greater contributor to NO_x in the upper troposphere than previously believed". Modelers have found that increasing the lightning-NO source substantially is necessary to bring model-calculated and measured NO_x during the INTEX-A campaign into agreement. This increased lightning-NO source is consistent with recent midlatitude field campaigns that indicate that lightning-NO emissions from storms with midlatitude characteristics are larger than lightning-NO emissions from storms with tropical characteristics [*Huntrieser et al*., 2008; *Ott et al*., 2007]. CMAQ simulations with and without lightning-NO emissions were run for the summer of 2004. The impact of lightning-NO emissions on upper tropospheric NO_x and ozone can be seen in Figures 3 and 4, which compare INTEX-A NO_x and ozone observations from Flight 12 with output from CMAQ simulations with and without lightning-NO emissions.

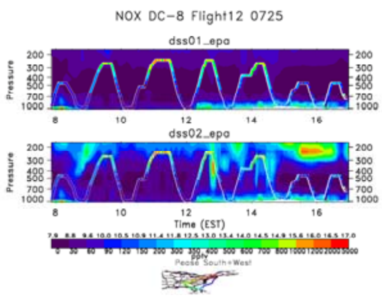

Figure 3. Curtain plot comparing CMAQ-calculated NO_x as a function of time (Eastern Standard Time) and pressure (hPa) with measurements from DC-8 Flight 12 on July 25, 2004. Top (bottom) panel shows results from simulation that did not (did) include lightning-NO emissions. Measured values are shown with a ribbon. Model-calculated values are shown in the background. The location and time of INTEX-A samples are shown on the US map. The color bar shows the time of one-minute average samples and the scale for the NO_x measurements

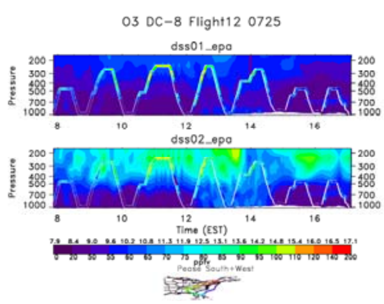

Figure 4. Same as Figure 3 but for ozone.

Clearly, lightning-NO emissions were responsible for a substantial portion of the ozone enhancement over the eastern United States on July 25. Similar enhancements were seen on several other INTEX-A flight dates (see Figures 5 and 6) although when averaged over all flight dates, CMAQ-calculated upper tropospheric NO_x remained more than a factor of two less than observed NO_x even with a 500 mole per flash lightning-NO source. This low-bias could indicate that the upper tropospheric lifetime of NO_x is too short in the model. The low-bias is also partly related to the lack of aircraft NO_x emissions in the model.

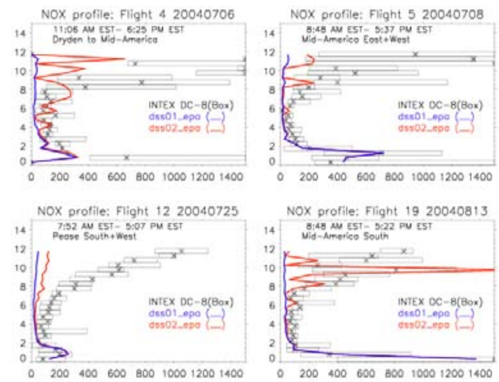

Figure 5. Mean NO_x profiles for INTEX-A Flights 4, 5, 12, and 19. Observed median mixing ratios within 0.5 km vertical bins are shown with an asterisk, while bars show the 10th and 90th percentiles. CMAQ-calculated median mixing ratios for simulation with (without) lightning-NO emissions are shown in red (blue).

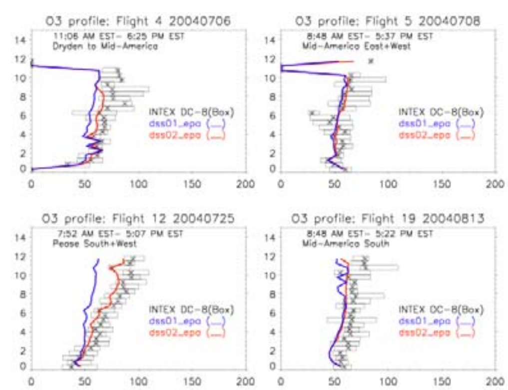

Figure 6. Same as Figure 5 but for ozone.

4. COMPARISON WITH SCIAMACHY NO_2 COLUMNS

Napelenok et al. [2008] found that low-biases in upper tropospheric NO_x in simulations without lightning-NO emissions made it difficult to constrain ground-level NO_x emissions using inverse methods and SCIAMACHY retrievals. Figure 7 compares CMAQ-calculated tropospheric NO_2 columns with SCIAMACHY columns. Overall, CMAQ calculated columns are 22% too low without lightning-NO emissions and 5% too low with lightning-NO emissions. With the exception of a north-south strip from eastern Nebraska to eastern Texas where biases increase slightly, the introduction of lightning-NO emissions, reduces biases throughout the US.

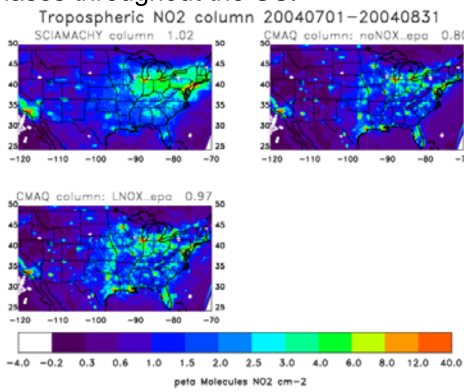

Figure 7. Mean tropospheric NO_2 column is shown for July 1 – August 31, 2004. Clockwise from upper left: SCIAMACHY column, CMAQ column from simulation without lightning-NO emissions, and CMAQ column from simulation with lightning-NO emissions. CMAQ columns were calculated using model output at the time of the ENVISAT overpass (10:00 AM LT). A tropopause pressure of 150 hPa is assumed. No averaging kernel is applied to model output. Only days with SCIAMACHY retrievals are used in calculating the CMAQ averages.

5. IMPACT ON SURFACE LAYER OZONE

Lightning-NO emissions contribute substantially to tropospheric NO_2; however, their contribution to surface layer ozone amounts is not large. Figure 8 shows the contribution of lightning-NO emissions to 8-hour maximum ozone during the July 1 to August 31 time period. The top panel shows the mean contribution while the bottom panel shows the contribution on the 95% ozone day during the July 1 to August 31 time period. Mean increases of 1-3 ppbv are typical.

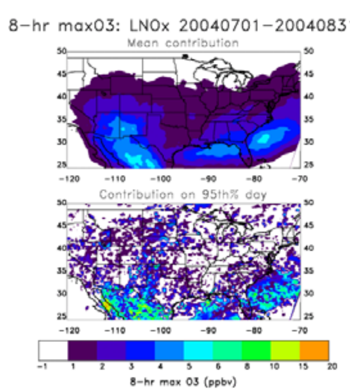

Figure 8. Contribution of lightning-NO emissions to 8-hour maximum ozone amounts are shown for the July 1 – August 31 time period. Top panel shows mean contribution during period. Bottom panel shows the contribution in each grid box on the day when the CMAQ simulation without lightning-NO emissions had its 3rd highest 8-hour maximum ozone in that grid box.

The mean contribution on high ozone days is a bit less than the mean contribution on all days. On days when 8-hour maximum ozone exceeded 75 ppbv for the CMAQ simulation without lightning-NO emissions, lightning-NO emissions contributed 0-1 ppbv of ozone on 42% of the days, 1-2 ppbv of ozone on 34% of the days, 2-3 ppbv on 12% of the days, 3-4 ppbv on 6% of the days, and 4 or more ppbv of ozone on 6% of the days.

5. SUMMARY

A lightning-NO parameterization scheme has been developed for CMAQ. The scheme assumes flash rates are proportional to the MM5 precipitation rate but then adjusts the flash rates locally and monthly so that flash rates best match NLDN-based flash rates.

The introduction of lightning-NO emissions leads to better agreement with INTEX-A profiles of NO_x and ozone, although CMAQ-calculated NO_x is still too low on most of the flights. The mean bias between CMAQ-calculated tropospheric NO_2 columns and SCIAMACHY columns is reduced from 22% to 5%. Lightning-NO emissions do not have a large impact on surface layer ozone amounts. Surface layer ozone amounts on high-ozone days increase by less than 3 ppbv due to lightning on 88% of high-ozone days.

6. REFERENCES

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