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Atmospheric Chemistry and Physics Discussions

The effect of lightning NO_x production on surface ozone in the continental United States

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Abstract

Lightning NO_x emissions calculated using the U.S. National Lightning Detection Network data were found to account for 30% of the total NO_x emissions for July–August 2004, a period chosen both for having higher lightning NO_x production and high ozone levels, thus maximizing the likelihood that such emissions could impact peak ozone levels. Including such emissions led to modest, but sometimes significant increases in simulated surface ozone when using the Community Multi-scale Air Quality Model (CMAQ). Three model simulations were performed, two with the addition of lightning NO_x emissions, and one without. Domain-wide daily maximum 8-h ozone changes due to lightning NO_x were less than 2 ppbv in 71% of the cases with a maximum of 10-ppbv; whereas the difference in 1-h ozone was less than 2 ppbv in 77% of the cases with a maximum of 6 ppbv. Daily maximum 1-h and 8-h ozone for grids containing O_3 monitoring stations changed slightly, with more than 43% of the cases differing less than 2 ppbv. The greatest differences were 42-ppbv for both 1-h and 8-h O_3, though these tended to be on days of lower ozone. Lightning impacts on the season-wide maximum 1-h and 8-h averaged ozone decreased starting from the 1st to 4th highest values (an average of 4th highest, 8-h values is used for attainment demonstration in the U.S.). Background ozone values from the y-intercept of O_3 versus NO_z curve were 42.2 and 43.9 ppbv for simulations without and with lightning emissions, respectively. Results from both simulations with lightning NO_x suggest that while North American lightning production of NO_x can lead to significant local impacts on a few occasions, they will have a relatively small impact on typical maximum levels and determination of Policy Relevant Background levels.

1 Introduction

Nitrogen oxides (NO_x=NO+NO_2) play a key role in tropospheric chemistry and impact the tropospheric ozone (O_3) budget. Sources of NO_x include fossil fuel combustion,

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biomass burning, soil release, oxidation of atmospheric ammonia, lightning, and stratospheric NO_x. Recent global estimates of NO_x production from lightning vary from 1 to 20 Tg (N)/yr (Martin et al., 2007; Labrador et al., 2005; Boersma et al., 2005), though 5±3 Tg N/yr is applied in most global modeling (Ridley et al., 2005; Schumann and Huntrieser, 2007).

Lightning can lead to significant increases in NO_x in the middle and upper troposphere and contribute significantly to columnar abundance of NO_2 (Choi et al., 2005; Zhang et al., 2003; Tie et al., 2002; Bond et al., 2001; Zhang et al., 2000; Pickering et al., 1998; Delmas et al., 1997; Ridley et al., 1996; Beirle et al., 2006; Martin et al., 2006). This source is particularly important in the Southeastern U.S. during the summertime (Biazar and McNider, 1995; Bond et al., 2001; Hudman et al., 2007). However, such emissions often are not included in regional modeling, unlike global modeling (Choi et al., 2005; Egorova et al., 1999; Stockwell et al., 1999; Tie et al., 2002; Allen and Pickering, 2002). With further opportunities for evaluating our models using satellite measurements which indicate the importance of lightning NO_x enhancements (Boersma et al., 2005; Choi et al., 2005; Christian et al., 2003), including such emission to regional models should be considered.

Variability in background surface level ozone over the United States is a critical issue strongly related to air quality policy and recent ozone criteria document (E.P.A., 2006) investigated such levels in terms of the Policy Related Background (PRB). PRB is defined as the distribution of O_3 concentrations that would be observed in the U.S. in the absence of anthropogenic (man-made) emissions of O_3 precursors in the U.S., Canada, and Mexico. Estimates of background ozone range between 15–45 ppbv (Altshuller and Lefohn, 1996; Fiore et al., 2002; Hirsch et al., 1996; Liang et al., 1998; Lin et al., 2000; Trainer et al., 1993), though some studies suggest that natural background ozone is higher than the 25–45 ppbv range recently used as the PRB. A higher PBR impacts how readily one can achieve the National Ambient Air Quality Standard (NAAQS) for ozone (Lefohn et al., 2001). The current NAAQS is 0.08 ppmv, calculated as the 3-yr average of the fourth-highest daily maximum 8-h average ozone concen-

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tration measured at each monitor within an area over each year. Recently EPA has proposed tightening the standard to 0.070–0.075 ppmv further increasing the importance of how ozone production from lightning might impact levels. Fiore et al. (2003) suggested the O_3 background is generally 15–35 ppbv, with occasional incidences of 40–50 ppbv at high-altitude western sites in spring and these cases would not compromise the current 80 ppbv U.S. standard. Here, impacts of lightning NO_x emissions on surface ozone and contribution to PRB are investigated using the MM5-SMOKE-CMAQ system (Byun and J.K.S., 1999; Houyoux and Vukovich, 1999; Seaman, 2000) applied to July–August 2004. CMAQ simulations can give additional information because the effect of lightning might lead to high ozone at finer scales which can not be captured using current global models. July–August 2004 is chosen for a variety of reasons. First, pollutant data are available not only from the routine, ground based monitors, but from satellites and airborne platforms as well [ICARTT-International Consortium for Atmospheric Research on Transport and Transformation, NASA INTEX-NA – Intercontinental Chemical Transport Experiment-North America, Mission 2004], allowing for more detailed evaluation and investigation. Second, this period had a number of ozone exceedances across the U.S. (2008b). Third, this period has frequent convective events and lightning NO_x formation. As such, July–August is a top candidate for identifying the potential impact of lightning NO_x formation on high ozone levels and non-attainment.

2 Method

2.1 Model description

The Community Multi-scale Air Quality Model (CMAQ) (Byun and Schere, 2006) with the SAPRC99 Chemical Mechanism (Carter, 2000) is used in this study. CMAQ is an advanced, atmospheric chemical transport model widely used for both research and regulatory applications. The modeling domain has a 36 km×36 km horizontal grid res-

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olution covering the continental United States, Southern Canada and Northern Mexico including 13 vertical layers reaching up to approximately 15 km. The Fifth-Generation NCAR/Penn State Mesoscale Model (MM5) (Seaman, 2000) is used to develop the meteorological fields, and the Sparse Matrix Operator Kernel Emissions (SMOKE) (Houyoux and Vukovich, 1999) is used to prepare input emissions for CMAQ. The emission inventory used for the year 2004 is projected from the emissions inventory for the year 2002 (VISTAS2002) prepared as part of Visibility Improvement State & Tribal Association of the Southeast (VISTAS) (MACTEC, 2005). Emission projection use growth factors from the Economic Growth Analysis System (EGAS) Version 4.0, and control efficiency data obtained from EPA for the existing federal control strategies which were in place in 2004. Additionally, actual NO_x emissions in 2004 from U.S. power plants (i.e. electricity generating units, or EGUs) which are obtained from the continuous emissions monitoring (CEM) database from EPA website (2008a) are integrated into the emission inventory.

MM5 is run with 34 vertical levels to produce highly resolved meteorological information. The Grell scheme (Grell et al., 1995) is used to simulate cumulus (cloud) dynamics. The Four-Dimensional Data Assimilation (FDDA) (Dudhia et al., 2005) with the Pleim-Chang planetary boundary layer (PBL) scheme (Pleim and Chang, 1992) and Pleim-Xiu Land Surface Model (Xiu and Pleim, 2001) are also used. Simulated ozone is compared with ground observations for performance evaluation (Fig. 1).

2.2 Lightning NO_x emissions

Typical emission inventories, e.g. U.S. EPA's 1999 and 2002 National Emission Inventories (NEI) do not account for lightning NO_x emissions. Here, observed lightning strikes (U.S. National Lightning Detection Network, NLDN) are correlated to MM5 results, and used to enhance the traditional emission inventory. Three simulations were performed, one without lightning emissions, one with lightning emissions distributed using a vertical distribution based on Pickering (2007)[1] for July–August 2004 and another for sensitivity purposes using an older profile where more NO_x is emitted near the ground for August 2004. Detailed analysis shows how including those emissions impacts simulated ozone, particularly with respect to whether this more complete treatment impacts regulatory applications of air quality models.

Lightning NO_x emissions are estimated by using U.S. NLDN lightning flash data. NLDN coverage includes the continental U.S. and extends 200–300 km off the coastline, though includes only the cloud-to-ground (CG) lightning flashes. The detection efficiency for that data ranges from 80% to 90% for events with peak currents above 5 kA varying slightly by region over U.S. and decreases gradually away from the coast (Cummins et al., 1998). As such, the actual number of flashes is either the same or more than the reported values. Previous studies (Bond et al., 2001; Price and Rind, 1993; Schumann and Huntrieser, 2007) suggested that the number of intra-cloud (IC) flashes is about 3 times that of CG flashes, and we adopt this ratio in this study. This value is also consistent with the results that are derived from satellite lightning measurements from the Optical Transient Detector (OTD) for 1995–1999 over the contiguous U.S. (Boccippio et al., 2001). Even though this ratio might be on the upper range for some regions, we chose to use it to conservatively estimate the maximum likely impact on ozone. The vertical profile used for allocation of NO_x emissions is a modified form of widely used vertical profile for mid-latitudes (Pickering et al., 1998). The "older" vertical profile is based on the assumption that IC flashes produce approximately 10 times less NO than CG flashes (Price et al., 1997). More recent studies suggest that the production of NO by an IC flash is approximately equal to that of a CG flash (DeCaria et al., 2005; DeCaria et al., 2000; Fehr et al., 2004; Ott et al., 2007). The new scheme puts much less NO_x in boundary layers versus the previous profile (Pickering, 2006). The difference in the amount of NO produced per flash for IC and CG flashes increases NO from IC flashes in the upper troposphere. As mentioned above, the orig-

[1] Pickering, K. E., Personal communication, 2007.

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inal vertical distribution is modified by removing a fraction of lightning mass from the surface layers (0–1 km) and re-distributing this amount evenly to the layers between 5 and 12 km (Fig. 2) (Pickering, 2006).

Using DeCaria et al. (2005) led to an average of around 500 moles on NO per flash for both IC and CG flashes (DeCaria et al., 2005; Pickering, 2006). This value is also used for FLEXPART (Cooper et al., 2006) and GEOS-Chem (Hudman et al., 2007) simulations over North America during Summer 2004 and good comparisons with aircraft data have been obtained. Even though this value is in the upper range for the literature (Schumann and Huntrieser, 2007), results obtained from this assumption will likely give us an upper limit for the effects, so we can be more certain about the potential effects of lightning NO_x on surface O_3.

To develop a spatially and temporally detailed NO_x inventory, the emissions estimates, along with lightning events were processed to obtain hourly and gridded lightning emissions. NO emissions from lightning were located into model layers according to the modified vertical profile scaled to the cloud top of the particular clouds at each CMAQ grid cell. Cloud occurrence (cloud cover) and cloud top are obtained from MM5 predictions at each CMAQ grid cell. Cloud occurrences are compared with locations of lightning events. Predicted cloud information and actual lightning data from NLDN are combined to allocate the lightning NO_x emissions. Allocation uses either (1) the actual grid's cloud information if its cloud cover is not zero or (2) averaged non-zero neighbor grids' cloud information if cloud cover is zero in the location of an observed flash. Occurrence of lightning where no cloud is predicted was very rare: 0.18% of the time on average, with a 1.37% maximum on a specific day for the episode.

3 Results and discussion

Lightning NO_x emission is a significant fraction of the total (Fig. 3), accounting for 30% of the total emissions for July–August 2004. This amount is higher than what would be found for an annual average because summer months are the most lightning active

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times of the year. The assumed NO_x production rate per flash and the IC to CG ratio also have a significant effect on this fraction and are still rather uncertain. The spatial distribution of the lightning NO_x emissions shows high intensity in three regions; the Southeast, Texas and its surrounding, and the Midwest (Fig. 4).

Model results using the baseline and lightning enhanced inventories found that including lightning NO_x typically had a small impact on domain-wide, maximum 8-h O_3, with more than 71% of the daily peak concentrations being affected by less than 2 ppbv (Fig. 5). There are only 2 days with a significant difference with around 10 ppbv being the maximum. Impacts on 1-h O_3 are similar, with more than 77% of the domain-wide daily maximum ozone concentrations being within 2 ppbv (Fig. 5). The largest impact is 6.4 ppbv. On average, the difference in domain-wide peak was 1.1 ppbv and 1.7 ppbv for 1-h and 8-h averages, respectively.

Further investigations into how lightning might impact ozone attainment in the U.S. were carried out with the model results at the 692 grid cells where one or more routine ozone monitoring stations are located. Daily maximum 1-h and 8-h O_3 with lightning emissions are generally, but not always, higher than the base case (Fig. 6). For grids with monitors, differences due to lightning ranged from –16.7 to 41.5 ppbv for 1-h O_3 and –6.4 to 42.4 ppbv for 8-h O_3. When the base model O_3 is either quite high (more than 100 ppbv) or low (less than 40 ppbv) the differences are smaller (Fig. 7). This shows that the maximum impacts do not occur when ozone is the maximum in the domain in cities where the anthropogenic emissions dominate. The distribution of the differences between two scenarios shows that for more than 43% of both 1-h and 8-h O_3, the difference is less than 2 ppbv (Fig. 6), though there are some extreme cases of more than 20 ppbv. This suggests that not including lightning NO_x emissions will typically, but not always, have little impact.

The simulated first, second, third, and fourth highest daily O_3 values for the simulation period are also compared at each of these 692 grid cells (Fig. 8). The impact of lightning on the 4-highest values of 8-h O_3 is smaller than for daily maximum values. The number of outliers where additional ozone coming from lightning is very high decreases and the correlation between the base case and lightning case ozone increases going from the 1st to 4th highest values (Fig. 8). Out of the 2768 (4×692) cases, a total of 219 (77 for 1st, 73 for 2nd, 36 for 3rd and 33 for 4th highest values) occurred where including lightning NO_x led to an exceedance of the NAAQS that was not simulated otherwise. Differences between the two cases narrows from the 1st to 4th highest values (Fig. 9) suggesting the effect of lightning on 4th highest values is less than on the highest values, and that significant impacts of lightning on peak ozone levels is infrequent. The maximum difference in the 4th highest values is around 14 ppbv whereas it is around 38 ppbv for the 1st highest value. 1-h O_3 follows a similar pattern as 8-h O_3 (Fig. 9) and the maximum difference in the 4th highest values is around 19 ppbv whereas it is around 36 ppbv for 1st highest value. While this work does find cases where including lightning NO_x leads to additional simulated exceedances of the NAAQS, the approach now used for demonstrating future attainment using a "Relative Reduction Factor" (RRF) will minimize the impact of lightning NO_x in such effects. RRF is the ratio between the modeled future year and basecase 8-h daily maximum O_3 concentrations at a given monitor averaged over the simulation period used for calculating the future design value by multiplying it with the current design value (E.P.A., 2005) The spatial distribution of the lightning effects in the 1st highest daily 1-h and 8-h O_3 shows that the most highly affected regions are high intensity lightning regions where biogenic VOC's are relatively abundant and O_3 production is limited by NO_x (Fig. 10).

A sensitivity simulation for August 2004 using the older vertical profile showed less O_3 on the surface than the modified profile even though it allocates the maximum amount of NO_x on the surface. Reasons for the depressed O_3 come from the timing of those emissions and the level of NO_x production. Much of the lightning-derived NO_x is due to strong convective activity and found later in the afternoon into the evening and night. Such periods have decreased photolysis from cloud cover or high solar zenith angles, and large amounts of fresh NO_x emissions do not immediately contribute to photochemical ozone production and can lead to ozone decreases by both direct titration and nighttime chemistry that further reduces ozone as well as depletes much of the NO_x produced (Russell et al., 1986). Accordingly, the sensitivity simulation resulted in higher daily maximum domain-wide O_3 values for some days, and lower on others depending upon the timing and extent of NO_x production. The destruction effect is more obvious if AIRS monitoring stations are investigated. These results from AIRS monitors are focused on urban areas, and as such have higher NO_x levels. Including lightning emissions in the sensitivity case results in somewhat greater decreases with few increases in the first, second, third and fourth highest values compared to the updated profile. Overall, the net additional exceedances from lightning with the older lightning NO_x distribution profile are either zero or negative for virtually all cases.

The effect of North American lightning NO_x emissions on average background ozone can also be investigated by finding the y-intercept of O_3 versus NO_z (Hirsch et al., 1996; Trainer et al., 1993) which gives the background ozone as 42.2 and 43.9 ppbv without and with lightning NO_x emissions, respectively (Fig. 11). The simulated difference, 1.7 ppbv, is statistically significant at the 95% confidence level. The slope of the line in Fig. 11 suggests overall ozone production efficiency (OPE) of about 5.47 for the base case and 5.66 for the lightning case showing that addition of lightning NO_x resulted in a decrease in overall OPE's for ground level ozone. As discussed above, injection of NO_x in the late afternoon and night increases NO_z, but can decrease ozone due to direct titration and nighttime chemistry.

4 Conclusions

Lightning NO_x emissions estimated in this study account for 30% of the final total U.S. emissions for July–August 2004 with the assumed NO_x production rate per flash and IC/CG ratio. Spatial distributions show emissions are significant in three main regions in the U.S.: the Southeast, Texas and surrounding and the Midwest. Resulting differences in daily maximum 1-h and 8-h O_3 were typically small, with more than 77% and 71% of concentrations being within 2 ppbv, respectively. On average, the difference in the domain-wide peak was 1.1 ppbv and 1.7 ppbv for 1-h and 8-h O_3 values. While

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infrequent, there were a few days with significant increases in the domain-wide peaks (up to 6 ppbv in 1-h and 10 ppbv in 8-h levels). However, these did not occur on days of high maximum ozone.

In the 692 grids with ozone monitors, daily maximum 8-h O_3 with lightning emissions are usually, but not always, higher than the base case. Differences in the grids with monitors ranged from –16.7 to 41.5 ppbv for 1-h and –6.4 to 42.4 ppbv for 8-h O_3. For more than 43% of the cases, the differences were less than 2 ppbv, but there are a few cases of more than 20 ppbv increases for 1-h and 8-h O_3. The 1st to 4th highest values for these grids experienced a smaller simulated impact. The correlation between base case and lightning case ozone increased going from the 1st to 4th highest values showing the effect of lightning on the 4th highest is less than on the highest. The maximum difference in the 4th highest ozone in the domain is around 19 ppbv (36 ppbv for 1st highest) for 8-h O_3. These results indicate lightning NO_x can cause significant increase of ozone in some extreme cases but its effect on ozone decreases if these extreme cases are excluded.

Allocating more NO_x on the surface with the sensitivity simulation for August 2004 showed less O_3 on the surface. The destruction effect is more obvious for AIRS monitoring stations which focus on urban areas. Overall, the net additional exceedances from lightning with the older lightning NO_x distribution profile are either zero or negative for virtually all cases.

Even though the North American emission of NO_x from lightning is significant, its impact on daily maximum 8-h O_3 for entire U.S. and policy relevant background ozone is typically small. There was only a 1.7 ppbv increase in regional background ozone. On the other hand, spatial distribution of these emissions and some occasions of higher ozone increases in our results suggest that these can lead to significant local impacts on a few occasions, especially in the southern U.S., and analysis of ozone exceedances should consider if lightning may be a contributing factor. Lightning elsewhere in the world may also contribute to background ozone over North America through long-range transport.

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Given the rather minor impacts of lightning NO_x on ground level ozone found here, reducing the assumed emissions would lead to a nearly linear reduction in the ozone impact calculated (Cohan et al., 2005).

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Table 1. The number and percentage of exceedances of 1st, 2nd, 3rd and 4th highest 1-h and 8-h maximum O_3 values for model grids with AIRS ozone monitoring stations for July–August 2004 with and without lightning. (prior O_3 NAAQS 1-h: 0.12 ppmv, 8-h: 0.085 ppmv).

Ozone		Base case exceedances		Lightning case exceedances		Increase/decrease due to lightning	
		Number	%	Number	%	Number	%
1-h	1st	37	2.67	41	2.96	4	0.29
	2nd	25	1.81	28	2.02	3	0.22
	3rd	22	1.59	23	1.66	1	0.07
	4th	18	1.30	20	1.45	2	0.14
	Total	102	1.84	112	2.02	10	0.18
8-h	1st	210	15.17	287	20.74	77	5.56
	2nd	150	10.84	223	16.11	73	5.27
	3rd	100	7.23	136	9.83	36	2.60
	4th	77	5.56	110	7.95	33	2.38
	Total	537	9.70	756	13.66	219	3.96

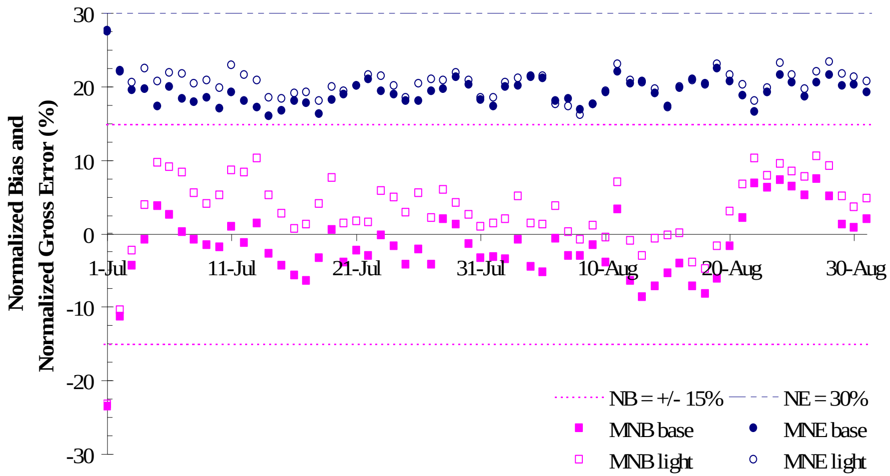

Fig. 1. Domain-wide daily normalized bias and error for O_3 in CMAQ when compared with AIRS, CASTNET and SEARCH O_3 measurements (for all pairs of O_3>40 ppbv).

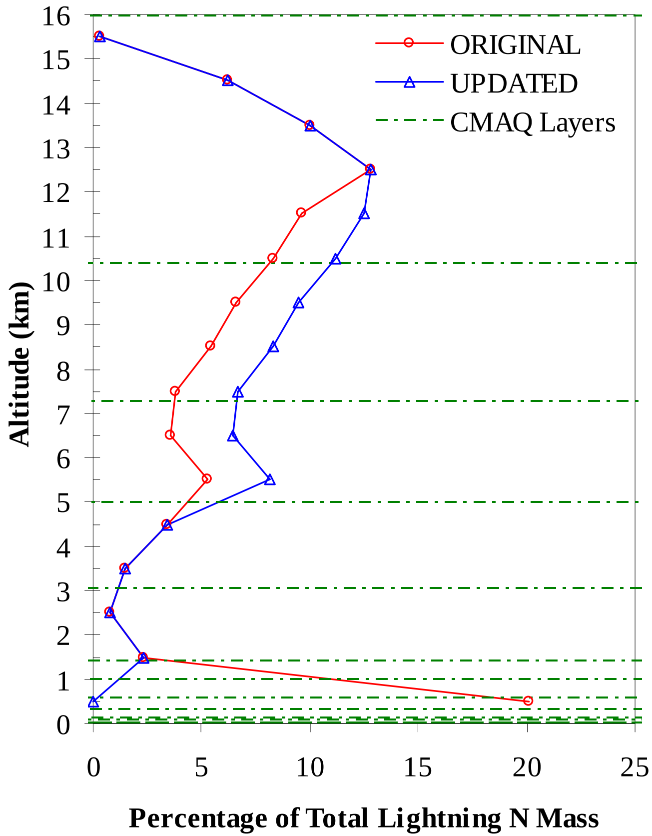

Fig. 2. Original and updated vertical profiles used for allocating lightning NO_x emissions into model layers.

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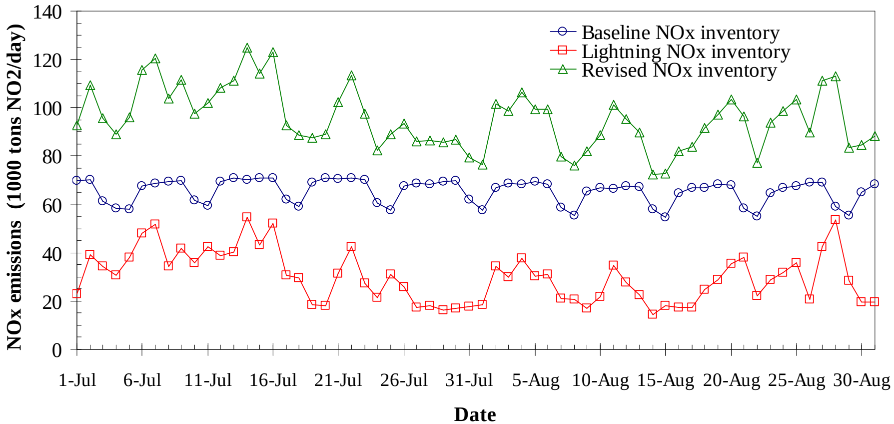

Fig. 3. Domain-wide baseline, lightning, and revised NO_x emissions (as tons NO_2/d) for July–August 2004.

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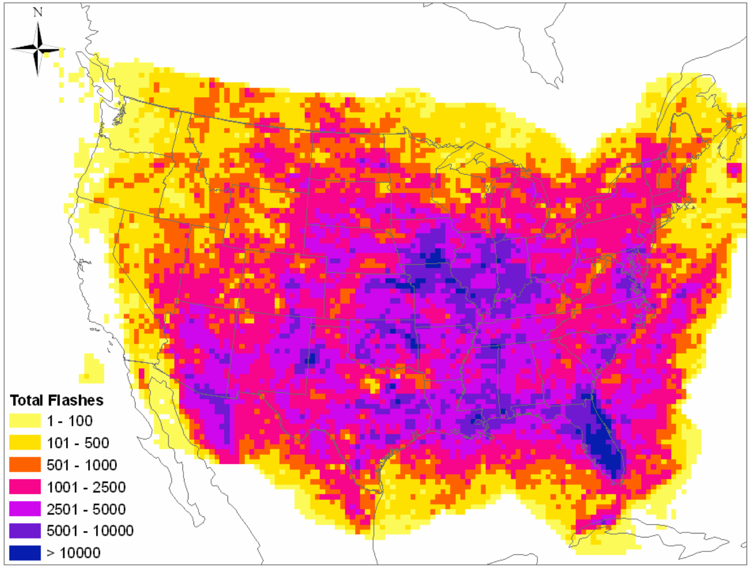

Fig. 4. Domain-wide lightning intensity map for July–August 2004.

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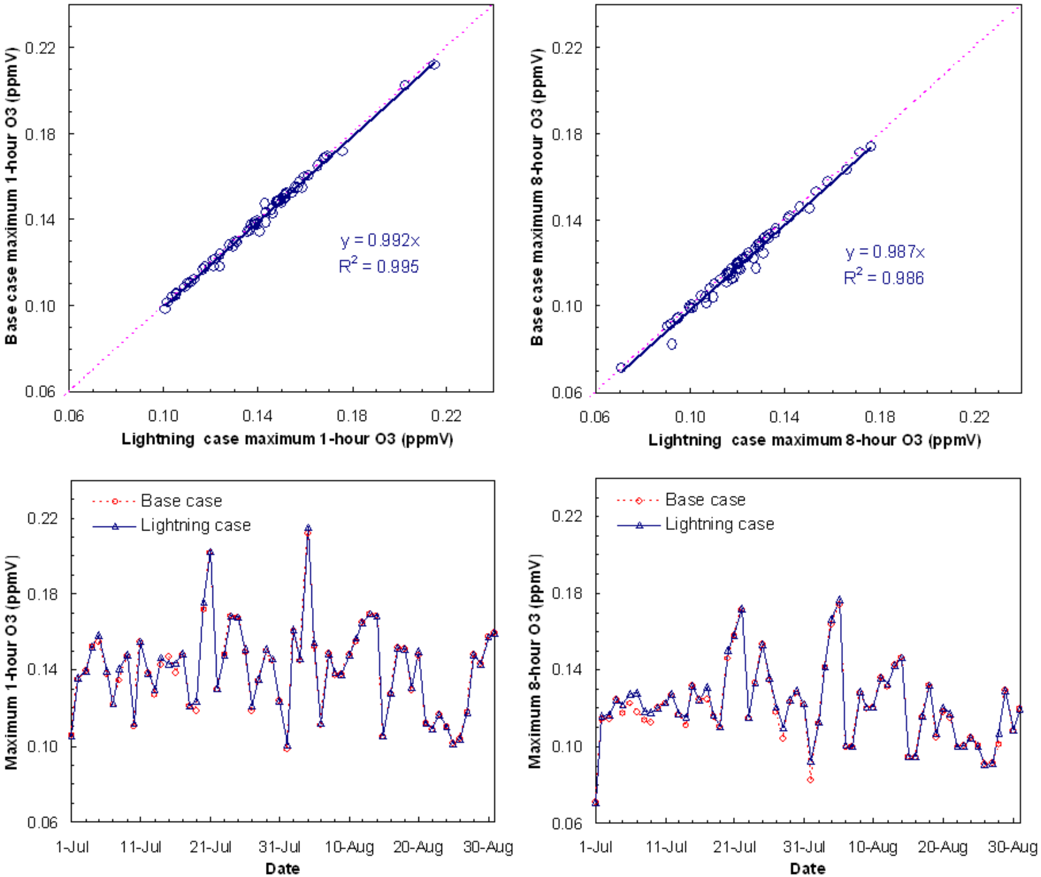

Fig. 5. Temporal evolution of domain-wide daily maximum 1-h (left panel) and 8-h (right panel) O_3 values with versus without lightning for July–August, 2004 (One grid with the maximum 1-h and 8-h O_3 is selected throughout the whole domain for each day).

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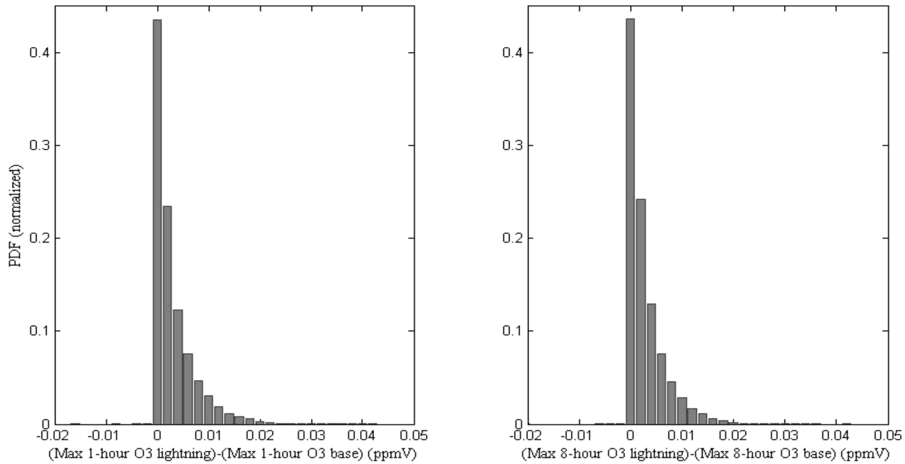

Fig. 6. Distribution of the change in daily 1-h and 8-h maximum O_3 values for model grids with AIRS ozone monitoring stations for July–August 2004.

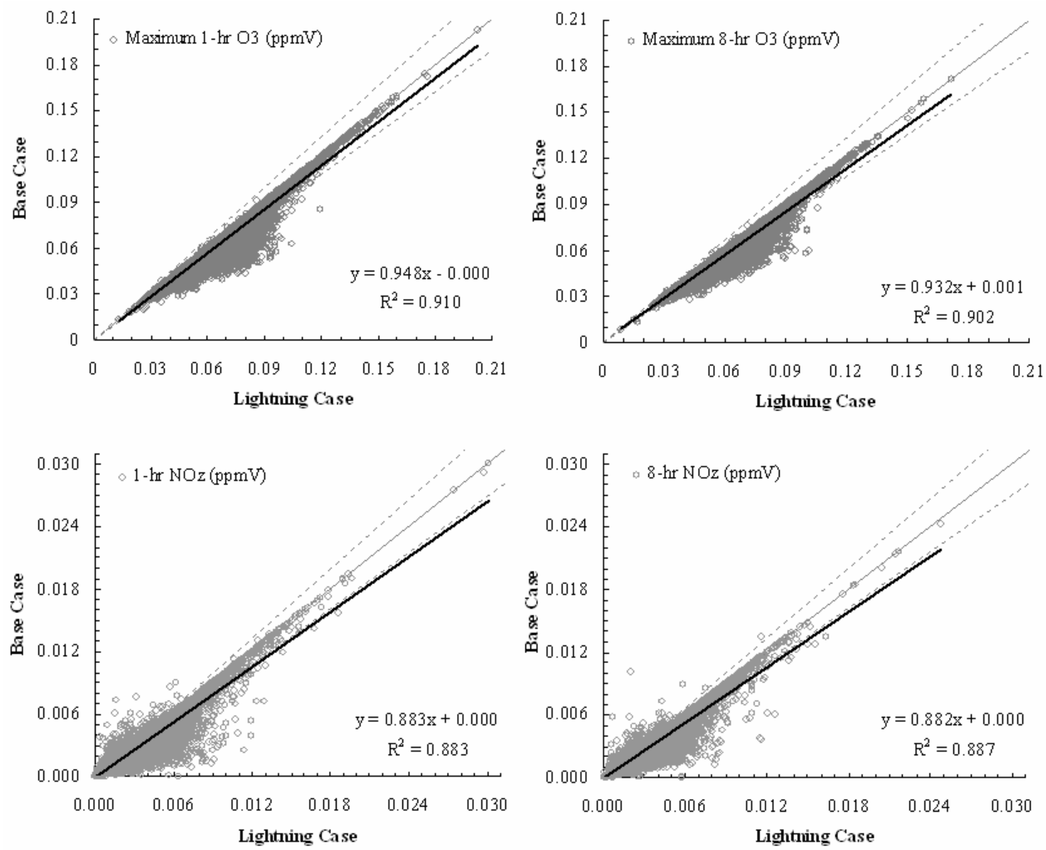

Fig. 7. Daily 1-h (left panel) and 8-h (right panel) maximum O_3 values and corresponding 8-h NO_z values for model grids with AIRS Ozone monitoring stations for July–August 2004 (692×31 points, red lines indicate 1:1 line and ±%10 lines.

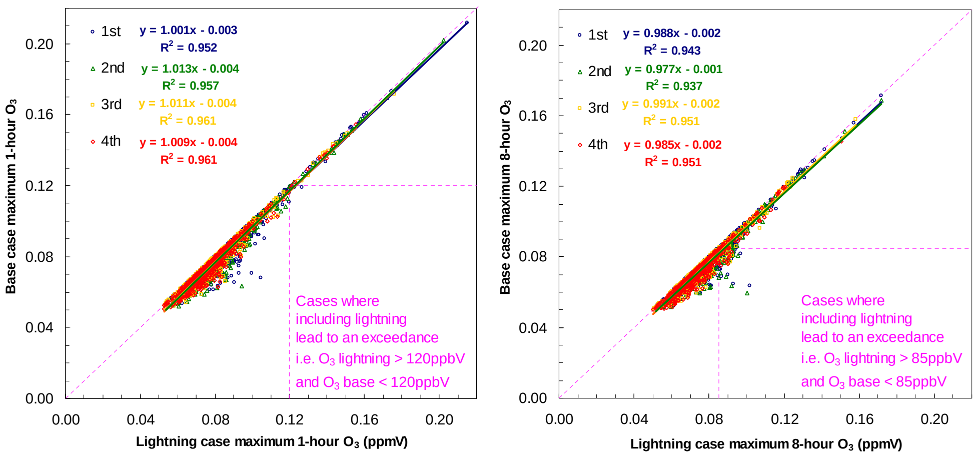

Fig. 8. The 1st, 2nd, 3rd and 4th highest 1-h (left panel) and 8-h (right panel) maximum O_3 values for model grids with AIRS ozone monitoring stations for July–August 2004. (2768 (692×4) points).

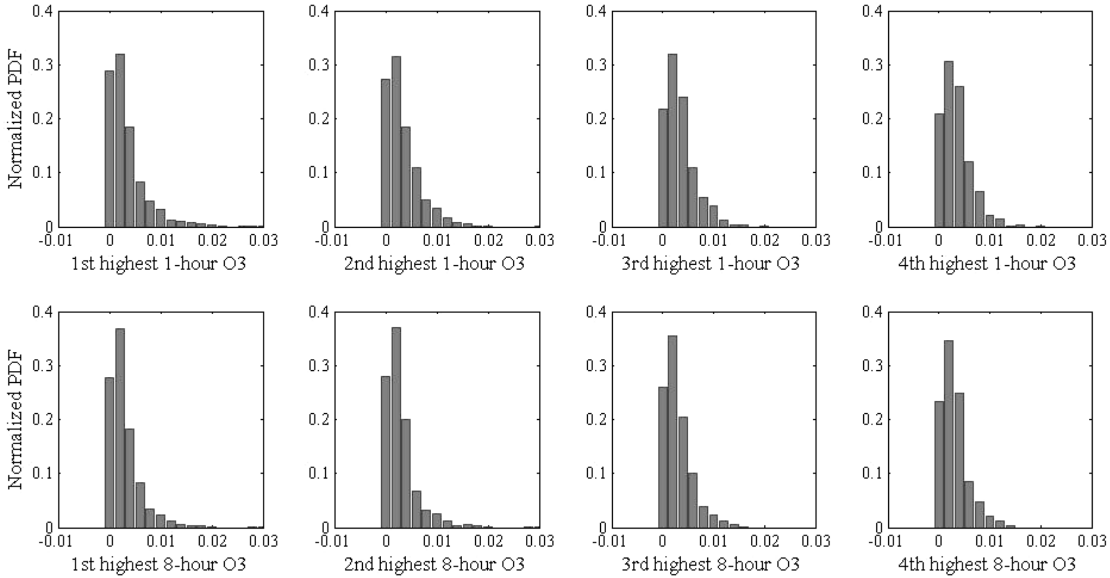

Fig. 9. Distribution of the difference (Lightning case-Base case) in the 1st, 2nd, 3rd and 4th highest 1-h (top) and 8-h (bottom) maximum O_3 values for grids with AIRS Ozone monitoring stations (PDF's are normalized and differences in units of ppmV).

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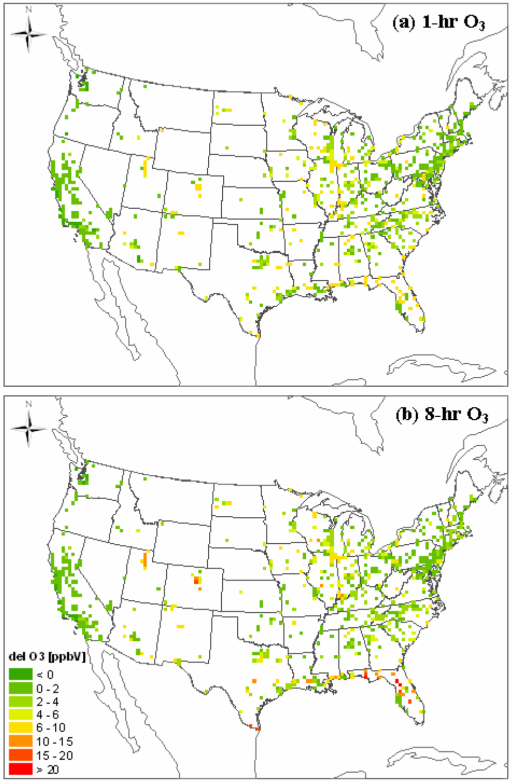

Fig. 10. Differences in **(a)** 1-h and **(b)** 8-h maximum O_3 values for model grids with AIRS Ozone monitoring stations for July–August 2004 (Difference in maximum daily first highest values are selected for each grid).

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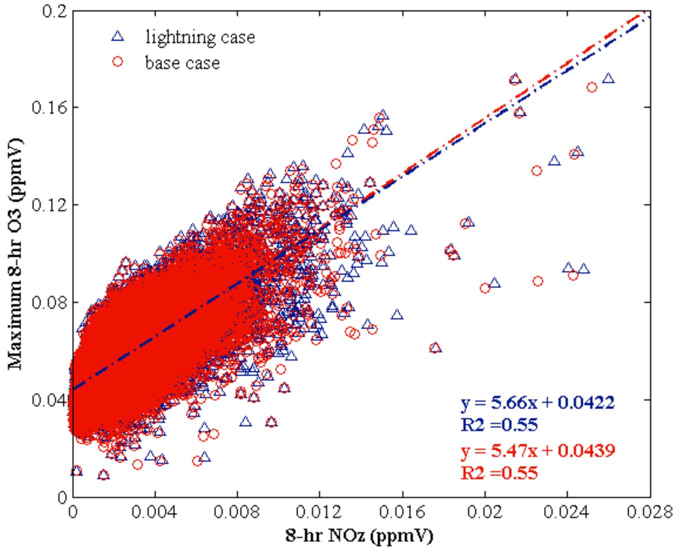

Fig. 11. Daily 8-h maximum O_3 versus corresponding daily 8-h NO_z (NO_y–NO_x) for grids with AIRS Ozone monitoring stations for July–August 2004. Shown are results for the base case (no lightning emissions) and lightning case.