Impact of lightning-NO on Eastern United States photochemistry during 2004 and 2006

Atmospheric Chemistry and Physics Discussions
General comments:

Anonymous Referee #1
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My major concern deals with the poor amount of new results in this paper that is much devoted to model development. There is however an interest for the CMAQ community with important model improvement. However most of the conclusions are derived (1) from the comparison of previous version of the model (without lightning) with the implementation of the lightning NO scheme (summer 2004) which hopefully gives better results than unrealistic simulations without lightning NO parameterization, and (2) after unconvincing evaluation of the model respect to in situ and satellite observations (NO2 and O3, summer 2006). The author should insist on the evaluation of the model respect to in situ and satellite observations. They may have a thorough discussion on the biases between the model and observations as they do well in Sec 3.5. It is indeed difficult to give general conclusions on the influence on lightning NO on tropospheric chemistry (Sec 3.3 Sec.3.4 and Sec.3.5) when the model is high biased. Moreover I found a lot of similarities in the methodology and the scientific objectives in a recent study of the authors, using another model (GMI): Allen et al JGR doi:10.1029/2010JD014062 2010. The only new results concerns in this paper concerns the discussion of urban versus rural sites for tropospheric NO2 columns in Sec3.1 and the impact on deposition of nitrogen species in Sec 3.4. I don't think this paper should be published in ACP unless important modifications and improvements.

Response to major comments from referee 1: From the author's general comments it appears that he/she has three major concerns with this paper.

1. The reviewer is concerned about the amount of new material in this paper. We believe there is easily enough new material to justify a paper. In this paper we discuss the causes of differences in NO_2 columns between urban and rural sites, the impact of lightning-NO on deposition of nitrogen species, and the sensitivity of upper tropospheric NO_x chemistry to uncertainties in model chemistry. These topics were not discussed in the Allen et al. JGR paper. Interest in these topics is not limited to the CMAQ community.

2. The reviewer is concerned about the high ozone bias in the model. The reviewer has a valid concern. We have spent considerable time diagnosing the causes of this bias. We have determined that the bias increases from the western domain of the model to the eastern domain of the model and is caused by excessive vertical mixing within CMAQ. The method of performing vertical advection in CMAQ is currently being changed by United States Environmental Protection Agency (EPA) modelers to reduce this problem.

An updated algorithm is expected to be released to the CMAQ community within a few months. However, as of October 2011, the new algorithm is still being tweaked and we are not able to present new results from it.

In addition to challenges in the ozone simulation, the satellite observations of ozone are challenging to interpret. The characterization of the model error is very different depending on which of the two ozone satellite products are used. Because of these problems we de-emphasized our analysis of upper tropospheric ozone in this paper. Specifically, we lessened the comparisons with OMI ozone fields and eliminated the comparisons with upper tropospheric TES data. Because the sonde measurements have relatively low uncertainty, we still show the sonde comparisons and use them to illustrate the problem and discuss the planned solution.

3. The reviewer believes the agreement between model-calculated and observed (in situ and satellite-retrieved) fields is not good enough to support robust conclusions.

As we state above, we will de-emphasize our ozone comparisons. In version one of this paper we presented three different satellite NO_2 products. Unfortunately, these products differed greatly and made comparisons with model output difficult to interpret. We now show two satellite products. We no longer show OMI AVDC columns because they are based on version 1 of the NASA standard product which has been shown to have a very large high-bias. A new version of the NASA standard product is expected to be released soon but is still unavailable as of early October 2011. We continue to show the DOMINO product because it contains an averaging kernel, which is critical for making comparisons with model output. Since version 2 of the DOMINO product is now available, we present it instead of version 1. We also show the DP-GC product, which is based on version 1 of the DOMINO product. After making these changes, the mean model columns and satellite columns agree to within -5 to +13%.

Abstract Lines 8-12 I am not sure the authors can give such conclusions on lightning NO contributions to NO2 tropospheric columns and O3 mixing ratios when the model is high biased in reproducing observations (Same remark for Sec 3.1, 3.2 and 3.4)

Yes, this statement does need to be qualified. We now state that the estimate is from a model with a lightning-NO source of 500 moles per flash. We have also removed the sentence where we state that lightning-NO reduces the model tropospheric NO_2 bias with respect to DOMINO from 41 to 14%. We now believe this reduction in bias cannot be estimated accurately as it is inappropriate to apply an averaging kernel to a simulation without lightning-NO emissions.

While the ozone high-bias is a concern, we estimate the contributions by taking differences between two model simulations. The differences are less sensitive to the high-bias than the actual values. In addition, to understand the impact of errors in the transported ozone on the CMAQ estimate of ozone production attributable to lightning NO, we designed a modeling experiment to examine the impact of ozone concentrations on ozone production in air masses influenced by lightning NO. Using observations from the INTEX-NA field campaign, we identified sixty-nine air masses that had recently experienced convection. The chemical and physical state of these air masses were used as initial conditions for a photo-chemical box model that simulates the production of ozone in each of these air masses for 48 hours following convection. We increased the ozone initial condition by 20% to quantify the impact of excess ozone in the CMAQ simulations. The net ozone production difference between the observed and perturbed air masses is show in this Figure. The sixty-nine air masses are segregated into four quartiles depending on their NO_x initial conditions. The box and whisker plot (Figure 1 of this response to reviewer) shows the range of results represented by the ensemble of air-masses in each NO_x quartile. For air masses immediately after convection with high NO_x levels (shown on the right), a 20% ozone error causes a 10% under-estimate of the ozone production over 48 hours. We have summarized these results in section 3.2 of the manuscript.

Introduction p 17703 lines 25 to 32 What is the interest of such sentence? Of course it is expected to have lower errors in a simulation with lightning-NO than without, as a simulation without lightning-NO is not realistic.

The interesting part of Napelenok's results is that low-biases in upper tropospheric NO_x contribute to biases in inverse-based estimates of surface-layer NO_x sources. It is important to remember that CMAQ was initially developed to serve the air quality community and upper tropospheric processes were considered secondary in importance. Low-biases in upper tropospheric NO_x due to the absence of lightning-NO emissions were not considered a major problem until it became clear that these low-biases were causing large errors in inverse-based surface layer NO_x emissions.

It would have been interesting to see how this new parameterization, compared to Koo et al., 2010 can better reproduce measurement during INTAS, not in comparison to a study without lightning NO from Napelenok et al. (2008)

We were not aware that Koo et al. were developing a lightning-NO parameterization when we began this study. Figure 2 of this response to reviewer compares the vertical distribution of lightning-NO used in this study with the July 2002 vertical distribution used in Koo et al. This figure was created by combining the NO emissions shown in Figure 4 of Koo et al. with information on the altitude of the model layers shown in Figure 4. This information was obtained from Koo et al. co-author Jeremiah Johnson. Clearly, Koo et al. place the emissions much lower in the model. Therefore if we were to simulate summer 2004 with their vertical distribution we would have an even larger low-bias in the upper troposphere.

Sec 2.3.1 Lines 10-14 p 17709 Similar lightning constraint approach from ground based and satellite observations has already been developed in previous studies that should be mentioned in the paper (Sauvage et al., 2007 with OTD-LIS; Jourdain et al., 2010 with NLDN). Moreover the author uses a very similar approach in a previous paper (Allen et al. 2010) with another model by constraining flash rates with OTD-LIS climatology and this should have been mentioned in the paper.
We have added the following to section 2.3.1:
Sauvage et al. (2007) and Allen et al. (2010) use time-averaged flash rates from OTD/LIS to constrain model flash rates. In this study, we follow a similar approach but use monthly average flash rates from the National Lightning Detection Network (NLDN) (Cummins et al., 1998) to constrain model flash rates. Jourdain et al. (2010) used daily average NLDN flash rates to constrain model flash rates.

lines 24-30 p17709 Description is unclear. " closely as possible " Please clarify. " to avoid very large flash adjustments " some precision should be added. Is this something happening a lot of times? How many adjustments are outside 0.1 and 10?
We now say the following:
Local adjustment factors ($\alpha_{i,j}$) are chosen so that when averaged over one-month periods of interest, model flash rates match observed total (sum of CG + IC) flash rates subject to the constraint that $\alpha_{i,j}$ is constrained to be between 0.1 and 10. During non-winter months, the lower constraints are invoked at approximately 7% of grid boxes within the 110°-70°W 25°-45° N analysis region, while upper constraints are invoked at approximately 2% of grid boxes. Therefore, monthly average model flash rates will not exactly match “observed” flash rates. Diurnal and day-to-day fluctuations in flash rates are not constrained.

lines 1-12 p 17710 Why are you using a different method to determine the percent of emissions for year 2004 and 2006? This is not explained and confuses the manuscript.
The method used to partition lightning-NO emissions in the vertical for the 2006 simulation was developed by Dr. William Koshak of NASA-MSFC under a recent research project entitled: NASA Lightning Results for Improving the CMAQ Decision Tool. Vertical profiles from this approach were not available when we began the 2004 simulation.

We now say the following:
In parallel with this study, vertical profiles of lightning-NO emissions for CMAQ were developed by W. Koshak of NASA-MSFC under a research project entitled: NASA Lightning Results for Improving the CMAQ Decision Tool. The 2004 simulations assume that emissions are proportional to pressure convolved by the mean April to September 2003-2005 vertical distribution of VHF sources from the Northern Alabama Lightning Mapping Array (Koshak et al., 2004; Hansen et al., 2010). The 2006 simulation uses more recent results from Koshak and assumes that emissions are proportional to pressure convolved by the segment altitude distribution of flashes from the same LMA (Koshak et al., 2010). With both approaches, NO_x emissions are distributed in all model layers from the surface to the layer containing the convective

cloud top. In practice, the difference between the approaches is small (Fig. 1) with the 2006 approach putting a bit less NO near the intracloud flash generated peak in the upper troposphere and a bit more NO near the peak in the mid-troposphere

The different methods should be tested for the same period (e.g. summer 2004) in order to evaluate the consequences on lightning NO simulations.
We believe the difference between the two methods is not large enough to warrant additional CMAQ simulations. We believe it makes the most sense to use the most recent scheme which is more physically based. We do not directly compare 2004 results with 2006 results.

Sec 2.3.2 p 17711 Could you please explain the important correlation differences between diurnal and daily variations in summer (and winter)?
Diurnal variations in flash rate are largest in the summer. This strong summertime signal is captured well by the model. Diurnal variations are more subtle in the other seasons and are not as well modeled.

During the fall through spring, most thunderstorms occur in the warm sector in advance of a cold front. In the summer, thunderstorms are more stochastic in nature and are difficult to model accurately.

lines 4-5 I don't see such a good agreement between the model and observations.
I now say the following:
This analysis showed that when averaged over a one-hour time period 35-45% of the strikes measured by the NLDN occur within WRF grid boxes with convective precipitation. When the averaging period is increased to one day, more than 90% of the strikes occur in WRF grid boxes with convective precipitation.

Coeffcient of determination would be more useful in such analysis (Rˆ2, not R) and is indeed quite low, except in summer for diurnal flash rate. Please clarify.
I will continue to use R as it is straightforward for the reader to calculate R^2 from R. Correlations between model and observed flash rates are often less than one might hope because of several factors. 1) Lightning is not always associated with convective precipitation. 2) The linear dependence of flash rate on convective precipitation rate is an oversimplification. 3) Model convective events, even when simulated, are often misplaced by a few hours or a few grid boxes. As noted before, diurnal errors are largest when diurnal forcing is weak (October – February) and day-to-day errors are largest when synoptic forcing is weak (summer).

line 25-28 What is a "stronger synoptic forcing"?
Summer 2004 was a cooler than normal over the eastern United States. The cooler temperatures were primarily the results of more frequent frontal passage (i.e., stronger synoptic forcing). We no longer use this terminology in the paper.

Sec3 A discussion should be realized on sensitivity test dealing with unrealistic simulations without lightning NO emissions (e.g. please see and add reference of Kun-

hikrishnan et al 2004) at least in order to interpret non linear processes such as for ozone

Kunhikrishnan et al.(2004) focused on photochemistry over the Indian ocean during 1997. They found that changes in ozone are not linearly proportional to changes in NO_x emissions and that nonlinear effects become important when NO_x emissions are perturbed by more than 30%. That study examines the sensitivity of ozone to NO_x emissions from different regions.

Our study has a different purpose – to calculate the quantity of ozone that exists because of NO produced from lightning. That is, we are interested calculating the amount of ozone that can be attributed to lightning, not the instantaneous change in ozone due to an NO_x perturbation. Exactly because ozone does not respond linearly to NO_x concentration, the best way to calculate the ozone due to lightning NO_x is with two simulations, one with and one without lightning NO production. This is the approach of several prior studies, for example Hudman et al. (2007), Sauvage et al. (2007b), Kaynak et al. (2008), Zhao et al. (2009).

We have added the following to the text:

The use of zero-out simulations to examine the amount of ozone that can be attributed to lightning is not strictly accurate as ozone production is a nonlinear function of NO_x concentrations (Liu et al., 1987). However, sensitivity runs have shown it is a good approach for determining the response of ozone to NO_x perturbations of less than 30-40% (Kunkrishnan and Lawrence, 2004), and it may be the best approach for determining the amount of ozone with a lightning-NO source (e.g., Hudman et al. [2007], Sauvage et al. [2007b], Kaynak et al. [2008], and Zhao et al. [2009]).

Sec 3.1 It is really hard to follow this section. The interest of this section is also very difficult to see as the methodology and the conclusions are very weak or confused, especially the ones on lightning NO contribution. The main interest of this section concerns the comparison between rural and urban regions and should be highlighted.

We agree that the conclusions in this section were weak. The primary problem was that the satellite-retrieved NO_2 data sets differed greatly making comparisons with model output ambiguous. As noted by the reviewers the section also suffers because some of the model/satellite comparisons are done without the use of an averaging kernel. We chose to show results without an averaging kernel because one was not provided with the NASA standard or DP-GC product.

We no longer compare with the NASA OMI standard product as it has been found to have a large high-bias. We have also replaced version 1 of the DOMINO product with version 2. These changes lead to better agreement between satellite products and with the model. This better agreement leads to stronger conclusions. In addition, we use averaging kernels wherever possible.

In order to highlight the rural/urban contrasts, we have reduced the amount of material in the rest of the section.

Fig.4 and Fig.5 demonstrates strong differences between model (with lightning NO) and DOMINO NO2 columns and it is hard to derive strong conclusions from a sensitivity test with and without lightning when the model is such biased compared to observations.

A sizeable portion of the model biases is due to problems with the observations. The NASA OMI AVDC product was derived from collection 1 of the NASA OMI NO2 standard product which is known to have a large high-bias in the summer time. In order to lessen the confusion caused by this highly biased product, we have removed this product from Figure 4. A new version of the DOMINO product has been released that is also more in line with the CMAQ output. We use this product in the updated plot. When these changes are made, the CMAQ column, averaged over this region of interest has approximately 5% low-bias with respect to v2 of DOMINO and a 13% high-bias with respect to the DP-GC product.

p17712 The domain used for averaging is big. Is there a strong geographical variability between the model and satellite to average on such a big domain?

Yes, the averaging domain is quite large. We chose a large domain to lessen the amount of day-to-day variability associated with variations in the location of valid OMI data due primarily to variations in cloud-cover. I'm not sure exactly what you mean by the second sentence. Are you asking if we needed to choose such a large region to get reasonable agreement? Not really, the agreement did not vary much with region size.

p17712 lines 3-4 What is the interest of such a sub-section if the comparison is not rigorous, as claimed by the authors? How conclusions on lightning-NO contribution can be deduced if the comparison is not rigorous, and when there is such a poor agreement between the model and the satellite retrieved column (except for mid July mid August with DP-GC mean columns)?

Figure 4 was intended to emphasize how different the satellite-retrieved products are. I did not apply an averaging kernel to the model output because one was not available for the DP-GC and AVDC products. I no longer show the AVDC product because of its high-bias and lack of an averaging kernel. I still show the DP-GC product. It does not include an averaging kernel but it is based on the DOMINO product and can therefore be compared to model output that has been processed using the DOMINO averaging kernel.

Where appropriate, I now process model output with an averaging kernel before comparing it to satellite-retrieved fields.

The agreement between satellite-retrieved and model columns is now reasonable, and the addition of lightning NO substantially reduces the model error.

Figure 5 Why the authors use the comparison between 3 different models column representations and the DOMINO columns only? Why not the others products?

I did not show the other products because they did not include an averaging kernel and I was attempting to be more rigorous here. An updated Figure 5 now includes panels showing the bias between model-calculated tropospheric NO_2 and the DOMINO and DP-GC columns.

Figure 6: p17714 lines 15-16 "addition of lightning NO decreases bias" A lower bias is at least expected when adding lightning NO. What is the new result of this sensitivity test?

I no longer address the change in bias due to lightning-NO emissions here, as it is not appropriate to apply an averaging kernel to a model simulation that has an unrealistic vertical distribution due to the absence of lightning-NO emissions. I now use this Figure (now Figure 5) to compare with both the DOMINO and DP-GC fields.

Sec 3.2 It is hard to derive conclusions on lightning NO contribution when the model gives better results without than with lightning NO emissions. Fig.9 shows indeed high biases between the best version of the model compared to observations. The strength between model and satellite given in Fig.9 are very weak (Rˆ2 does not exceed 0.6).

I was not surprised by the modest correlations, which are generally around 0.5. Even if the model was perfect, correlations are unlikely to be robust because the retrieval of tropospheric ozone from OMI is more of an art than a science. That said, the high-bias is real and is being addressed by sensitivity runs. We have removed the comparison with OMI time series from the paper as Figure 8 is sufficient to show that the model has a high-bias.

Figure 10: It is not because adding lightning in the model increases ozone by 6 to 12 ppbv, that the authors can conclude lightning NO production adds 6-12 ppbv to upper tropospheric ozone. Indeed the model does not match with satellite observations so it is hard to believe the simulations, and second non linear processes should be taken into account in such sensitivity tests (LNOx or NO LNOx) as ozone production (destruction) is not a linearly dependant of NOx (see the discussion in Kunhikrishnan et al GRL 2004).

Due to the high-bias in the model, we no longer show this comparison with TES. However, we still give model-based estimates of the lightning-NO contribution to upper tropospheric ozone at IONS sites. The values are obtained by differencing the results of simulations with and without lightning-NO emissions and should not be too sensitive to a high bias in model ozone. We will emphasize that these estimates are from a model. We have also performed box model calculations to ascertain the impact of the high ozone bias on our conclusions (see section 3.2).

Sec 3.3 Same remark than before on the conclusions related to the lightning NO impact on tropospheric column. The conclusions should be less affirmative.

OK. We now emphasize the magnitude of the lightning-NO source and that the conclusion is for the CMAQ model.

Sec 3.4 Fig.14 clearly shows that adding simulation with lightning NO improves a bit comparison with observation (for West America). However the comparison

between the model and observations are very low (R^2) and it is hard to conclude on the effect of lightning NO on the deposition of nitrogen species.
Correlation coefficients for the western United States are not really that bad equaling 0.62 for simulation noL and 0.70 for simulation LNO_x. Correlations exceed 0.85 (0.87 for noL and 0.89 for LNO_x) when corrections are made for biases in model precipitation (see Table 3). Given the uncertainties associated with scavenging parameterizations, we were quite pleased with the results.

Sec 3.5 I find this section the most interesting in the comparison and discussions between observations and model biases.
OK, we will leave this section mostly unchanged.

Minor comments
p 17704 line 10 " NASA OMI Aura Validation Data Center NO2 time series product ": please add reference
We no longer use the AVDC product as it is derived from the highly biased NASA standard product.

p17704 line 23 " that include negatives " I don t understand the sentence, please clarify.
As you likely know, tropospheric NO_2 columns are obtained by subtracting off stratospheric tropospheric NO_2 columns. Occasionally, the DP-GC algorithm calculates negative tropospheric columns for locations with small amounts of tropospheric NO_2. These values are unchanged in the DP-GC product with negatives but are set to zero in the DP-GC product without negatives. These negatives usually disappear when averaging is performed and a level 3 product created. In order to avoid confusion, we will remove “that includes negatives”. We will only show results from the standard DP-GC product, which includes negative values.

What is the incidence of using a NO2 column product that excludes scenes with cloud fraction higher than 30% (NASA time series) and another with cloud fraction higher than 50% (DOMINO)? An equal threshold value should be used both for NO2 and O3 retrieval product for comparison with model simulations (50% as mentioned for O3 in p 17705 line 50%)
These products were produced by different groups. Boersma et al. (DOMINO data product v2.0 users manual available at
http://www.temis.nl/docs/OMI_NO2_HE5_2.0_2011.pdf) suggest that users filter out all retrievals with cloud radiance fractions in excess of 50%. Celarier and Retscher (OMINO2e data product read me file available at
http://toms.gsfc.nasa.gov/omi/no2/OMNO2e_DP_Readme.pdf) produce a level 3 product that filters out all retrievals with geometric cloud fractions in excess of 30%. We used these recommendations in our first version of the paper. In our revised manuscript, this distinction is mostly moot as the NASA OMI standard product is no longer used. However, in anticipation of these comparisons, we now apply the geometric cloud fraction-based gridding scheme of Celarier and Retscher to the DOMINO product, while continuing to filter out retrievals with cloud radiance fractions

in excess of 50%. When the version 2 NASA product becomes available, we will follow the same gridding approach.

Sec 2.3 p 17707 Please describe the time period of the simulation (2006 and 2004). It is difficult to understand when start and end each simulation, whether or not a spin up is used.
All simulations had 10-days spin-up time. For example, the 2006 simulation was initialized on 22 December 2005. Section 2.3 has been updated to state this explicitly.

Sec 2.3 p 17706 line 27 Please add reference for the Kain-Fritsch parameterization and the WRF model, and after for the BEIS inventory, SMOKE .
Done

Line 13-15 p 17707 " prior work has shown CMAQ underestimates " Which work? without lightning NO emissions? Please clarify and add reference.
Yes, the Napelenok et al. (2008) study.

References Please check references. There is a clear need to add references to mention previous studies in the paper. Some of the references cited in the text do not appear in the Reference section.
We’ve gone back and cross-checked the references. We’ve added other references where appropriate.

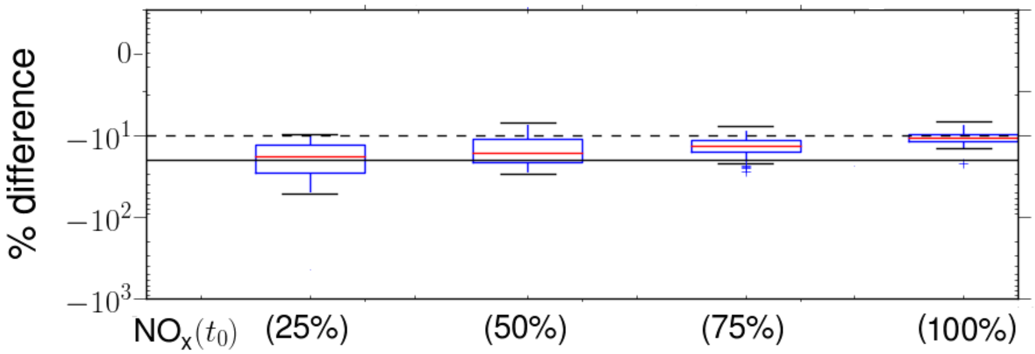

Figure 1. Box and whiskers show the percent change in ozone production when background ozone amounts in a box model initialized with observations from INTEX-A are increased by 20%. Results are shown for four NO_x quartiles determined by background NO_x amounts: low (25%), moderately low (50%), moderately high (75%), and high.

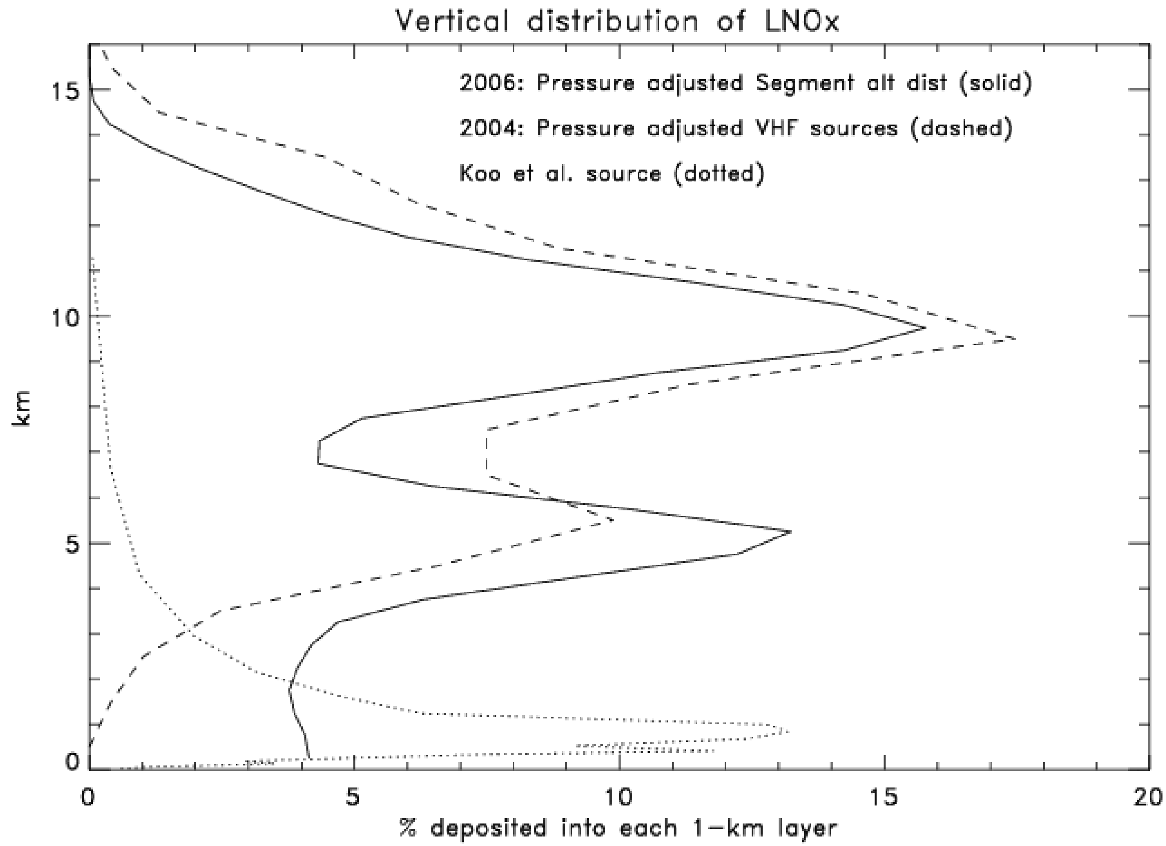

Figure 2. Vertical distribution of lightning-NO production assumed for CMAQ simulations by Allen et al. (2011) and Koo et al. (2010). The solid (dashed) black line shows the Allen et al. distribution for 2006 (2004). The dotted line shows the Koo et al. distribution.