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

Atmospheric Chemistry and Physics Discussions
General comments:

Anonymous Referee #3
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The authors have developed a "new lightning nitrogen oxide algorithm" in the CMAQ model and have used it to evaluate the impact of this source of NOx on tropospheric photochemistry over the USA during summer 2004 and 2006.

Major comments:
The paper does not provide a substantial contribution to the research on the source of NOx by lightning, even if this paper is certainly interesting for the CMAQ community.
We erred in calling this a "new lightning nitrogen oxide algorithm". It is similar to the one we presented in Allen et al. (2010). The focus of this paper is not the lightning algorithm. We assume a lightning-NO source (500 moles per flash), constrain monthly average convection-based flash rates with NLDN-based flash rates and then determine the impact of the resulting lightning-NO emissions on tropospheric photochemistry and nitrogen deposition over the eastern United States.

1) Indeed, the lightning–NO algorithm has already been introduced in CMAQ in Koo et al. (2010). Here, it only differs in that the emissions are scaled locally on a monthly basis using NLDN flash rates. The influence of this scaling on the results in term of NO2 mixing ratios is not really discussed in the paper.
We were not aware that Koo et al. were developing a lightning-NO parameterization when we began this study.
Koo et al. assumes emissions in a model layer are proportional to the product of the layer pressure times the convective cloud depth times the convective precipitation. They sum this product over a year and then scale the resulting sum to obtain 1.06 Tg N year-1 over North America. This value was obtained by multiplying the climatological NLDN CG flash rate (30 million flashes) by an estimate of the IC/CG ratio (2.8+1) , and a high-end estimate of the emissions per flash (667 Moles per flash).
Figure 1 of this response to reviewers 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.

Due to the very different partitioning of LNO_x emissions, we believe the impact of local scaling on model NO_x cannot be studied via a comparison of the two schemes. Figure 2 does show the difference in emission location between simulations with and without local scaling.

2) In addition, one goal of the paper (cf. Introduction) is to investigate whether introducing a source of NO from lightning in the CMAQ model would lead to better simulate NO2 concentrations in the upper troposphere. Of course, by adding a source of NO in the upper troposphere, we expect a decrease in the bias between measurements and model.
As expected, adding LNO_x improves the agreement with satellite-retrieved NO_2 columns and with NO_2 profiles.

Adding LNO_x does not improve the agreement with satellite-retrieved ozone columns or with eastern United States ozone profiles. 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 to eliminate or at least lessen this problem. An updated algorithm is expected to be available 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.

Because of these problems we will de-emphasize our analysis of upper tropospheric ozone in this paper. Specifically, we will remove the comparisons with OMI time series and TES data. We will still show the sonde comparisons and use them to illustrate the problem and discuss the planned solution. We have also performed box model simulations to investigate the impact of a high-bias in background ozone on upper tropospheric ozone production for varying amounts of NOx. Results are summarized in section 3.2.

3)Furthermore, the evaluation of model performances presented in the paper is not conclusive.
- For instance, the authors write that "Over the United States, LNOx is responsible for 20%-25% of the tropospheric nitrogen dioxide (NO2) column". I do not think the authors can state this. Indeed, the CMAQ model underestimates NO2 columns and NO2 mixing ratios in the upper troposphere as shown by the comparison with OMI and aircrafts measurements.
We will soften this conclusion by prefacing it with " For a LNO_x source of 500 moles per flash, model simulations indicate that …".
The mean model-calculated tropospheric NO_2 column was much lower than the mean NASA standard product column; however, the standard product column has been shown to have a large high-bias and is no longer used in this paper. Version 2 of the DOMINO product is also about 10% lower than version 1 of the DOMINO product. After these adjustments, the mean model NO_2 column is within -5 to +13% of the mean satellite-

retrieved columns (DOMINO and DP-GC). Of course, local differences can be much larger.
Of course, a large upper tropospheric low-bias remains with respect to INTEX-A measurements.

Furthermore, the authors do not take into account the source of NO by aircraft in their model. (In addition, owing to the non-linearity of the chemistry, comparing a simulation with NO from lightning and another that does not include NO from lightning, does not give the contribution of the lightning NO source to the NOx field). I think the authors should change their title.
Aircraft emissions were included in one of our 2004 simulations. They increase the mean column over the region of interest by only a few percent.

The purpose of our study is to calculate the quantity of ozone that exists because of NO produced from lightning. That is, we are interested in 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 NOx concentrations (Liu et al., 1987). However, sensitivity runs have shown it is a reasonable approach for perturbations less than 30-40% (Kunhikrishnan et al., 2004), and it has been used in prior studies including Hudman et al. (2007), Sauvage et al. (2007b), Kaynak et al. (2008), and Zhao et al. (2009).

The remark in the abstract "most of the differences between modeled and satellite-retrieved urban to rural rations are likely a consequence of the horizontal and vertical smoothing inherent in columns retrieved by OMI" should not be a result of the study. Indeed, it is expected.
The causes of differences between urban and rural biases are an area of active research. They may be an artifact of smoothing but could also indicate some real problems with tropospheric NO_y chemistry.

We now say the following in the abstract:
Differences in urban/rural biases between model and satellite-retrieved NO_2 columns were examined to identify possible problems in model chemistry and/or transport. CMAQ columns were too large over highly urban areas. Biases at other locations were minor after accounting for the impacts of lightning-NO emissions and the averaging kernel on model columns. These processes had a relatively large impact on the ratios indicating that the horizontal and vertical smoothing inherent in OMI-retrieved columns must be considered in model/satellite comparisons.

To overcome this problem, all the comparisons between model and satellite products should be done by taking into account the OMI averaging kernel. This is not always the case in the paper (figure 4 for instance).
Yes, in general, comparisons should be done after processing model output through an averaging kernel; however, an averaging kernel was not provided with the OMI standard product or the DP-GC product. We now process model output through the DOMINO averaging kernel before showing it in Figure 4. We no longer show the NASA standard product due to its high-bias and lack of an averaging kernel. We continue to show the DP-GC product as it is closely based on the DOMINO product.

In addition, the authors use different OMI products, the reading of their features is a little bit tedious. It would be helpful to summarize the information in a table. I think that errors on OMI columns should be added and used in the discussion.
We no longer show results for the AVDC product eliminating the need for a table.

By adding a source of NO from lightning, the authors scale up the ozone in the simulation of 2006 and increase the bias between the model and the observations. I agree that it is not necessarily due to the treatment of the lightning NO source in the model. But the authors do not investigate enough the reason for the discrepancies on ozone. They could give an estimation of the ozone bias in the fields (from GEMS and GEOS-CHEM) used for the boundary conditions. I would recommend improving the boundary conditions.
As stated above, we have spent considerable time diagnosing the causes of this bias and believe excessive vertical mixing within CMAQ rather than a bias in the GEMS boundary conditions is the primary cause of the problem. It is beyond the scope of this paper to fix this problem in vertical mixing. The upper troposphere ozone error is more important for our calculations of the contribution of lightning-NO to upper troposphere ozone. We have addressed this issue via box model calculations that are summarized in section 3.2.

To conclude, I think the authors should find what we can really learn from their study and build the paper around that instead of presenting a list of not very conclusive results.
Possible ideas: Do you find any influence of lightning events in OMI data? Percent of NO2 column due to lightning (roughly: LNOx –noL) vs NO2 column (LNOx) (when OMI avgk applied) would give ideas on the influence of lightning on the OMI column and if it is larger than the OMI errors. If yes, you could maybe focus on events before generalizing to United States and the whole summer?
Yes, a lightning-NO signal can be seen in OMI for certain events, especially over regions where anthropogenic emissions are not too large. A study of individual events is ongoing. It will be a separate study by itself and is beyond the scope of this paper.

We show the percent of the NO_2 column due to lightning in Figure 5. This estimate is based on CMAQ alone and is only appropriate for a 500 mole per flash source. We did consider determining the percentage as you suggest; however, the mean column is sensitive to the assumed vertical profile and that differs greatly between simulations with and without lightning-NO emissions. Therefore, determining the percent contribution of lightning-NO to the column using this approach is flawed and will lead to a high-bias.

Specific comments:

Abstract:

P17701, L26 please state here which uncertainties in the chemistry you will investigate in the paper.

Uncertainties in NO_y chemistry. This will be made clearer in the abstract and text.

Introduction:

I think the introduction section should be better organized. The overall context and prior work with the CMAQ model are mixed. The goals of the study should be better defined.

The overall context is now given in the first paragraph before the CMAQ model is introduced. The last paragraph of the introduction has been rewritten to emphasize the goals of this study. We have also shortened our discussion of other parameterizations as this information is either extraneous or of more use later in the paper.

P17702, L25 TES stands for?

Tropospheric Emission Spectrometer (TES). We no longer compare with data from this instrument.

P17703, L25 we expect to better represent NO mixing ratios in the upper troposphere when NO from lightning are included (see major comments above).

We will rewrite this section to emphasize that the 2004 simulations were performed to examine the maximum impact of uncertainties in tropospheric NO_y chemistry on upper tropospheric NO_2. As the reviewer notes, the fact that the model does better with LNO_x is not particularly surprising or noteworthy.

Section 2.1:

P17704 L23, "negatives" could you 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 DP-GC standard product, which includes negative values.

Why do you exclude pixels with cloud fraction higher than 50% for the NASA dataset and pixels with cloud fraction higher than 30% for the other dataset? you would need to say how much this has an impact on the difference between the two NO2 columns.

This question is mostly moot as we no longer compare with the NASA data set. 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) filter 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, we 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 new standard product data sets become available, we will use the same threshold and gridding scheme for them too.

You maybe need to say here how you compare NO2 columns from CMAQ with the different NO2 products from OMI. A table summarizing the features of the different OMI products would be helpful.
We have added several lines discussing how the DOMINO fields are mapped onto a level 3 grid and what is done with the CMAQ fields. We have also added a bit more information on the DOMINO and DP-GC products.

Section 2.2:
P17706, L1, change to ''The TES instrument is an infrared Fourier transform spectrometer with a spectral resolution of 0.1 cm-1 and a spectral range from 650-2250 cm-1 (Beer et al. 2001)" You can mention Worden et al. (2007) along with Nassar et al. (2008).
Due to the high-bias of the model, we no longer compare with the TES retrievals.

Section 2.3: Could you give a reference for SMOKE version 2.6?
Done, although the best reference is probably a link.

Could you describe how long the simulations last, when they begin and end?
All simulations had 10-days spin-up time. For example, the 2006 simulations were initialized on 22 December 2005 and ran through 31 December 2006. The 2004 simulations included the period from May 21, 2004 to August 30, 2004.

Why the boundary conditions are constant in time for the 2004 simulation? This should introduce errors in your simulations. Please explain why it is acceptable to use these boundary conditions.
We used fixed boundary conditions in 2004 to be consistent with Napelenok et al. (2008). Their results were one of the motivating factors for this project. We don't believe the use of fixed boundary conditions have a substantial impact on our conclusions with respect to the INTEX-A / model comparisons.

Section 2.3.1: Why do the authors do not scale the flashes to NLDN on a daily basis?

I assume you mean replace monthly-local scaling by daily-global scaling. Daily-local scaling could lead to a large mismatch between the location of model convection and the location of model lightning-NO emissions. Daily-global scaling is the approach followed by Jourdain et al. (2010) and one we considered. An advantage of this approach is that day-to-day variations in the magnitude of lightning-NO emissions are better constrained possibly leading to a better simulation of day-to-day fluctuations in tropospheric NO_2 column. A disadvantage of this approach is that daily scaling factors cause temporal fluctuations in emissions per flash. One difficulty with daily scaling is that NLDN data are occasionally unavailable. In addition, routine access to these fields would have to be obtained and possibly purchased from Vaisala. Since our goal was to create a method that can be used by the entire CMAQ community we decided against that approach for now.

Section 2.3.2: The title should be changed; it is not the evaluation of LNOx but the evaluation of Flash rates.
Done

P17711, L1, theses results are shown in the paper?
No, these results are not shown in a model figure. I will make this clear.

what should we conclude from
L1-L3 ? Simulated and observed daily variation of flash rate in summer do not agree, this has to be kept in mind when analyzing NO2 columns.

It shows that the agreement between model convective precipitation and NLDN lightning is so-so on an hourly basis but much better when averaged over a day. I have also added the following to the text.
During the fall through spring, most thunderstorms occur in the warm sector in advance of a cold front. Day-to-day variations in the locations of these storms are well captured with correlations averaging 0.80 and ranging from 0.67 to 0.90. In the summer, thunderstorms are more stochastic in nature and are difficult to model accurately. Observed and modeled daily-total flash rates are only weakly correlated during this period. The low correlations during this time period mean that the simulation of day-to-day variations in summertime upper tropospheric NO_2 is unlikely to improve when lightning-NO is added to CMAQ.

P17711, L27 what is this stronger synoptic forcing in 2004 ? the agreement is better in Aug 2004 but not in July 2004. Please could you clarify what we can learn from this section and would be important to understand the NO2 comparisons?
Yes, I think I'll eliminate that synoptic forcing statement. As a whole, the summer of 2004 had more frontal passages through the center of the United States than a typical year. However, it may be a stretch to say that that is the cause of the differences between 2004 and 2006.

Section 3.:

P17712, L7 can you explain why do you perform a simulation airLNOx? Your simulation LNOx do not have aircraft NO emissions?
Correct, we only included LNOx emissions for one sensitivity run during 2004. We will make sure this is clear in the model description. Their impact on mid- and upper-tropospheric NO_2, NO_x, and O_3 can be seen in the last column on Table 5.

Section 3.1:
Figure 4: I have a problem with this figure. You say that the comparison is not rigorous because you did not adjust the model output with the averaging kernel. I think you should not show this comparison between the model and OMI without adjusting the model results. You can not compare CMAQ and domino either because they not are at the same horizontal resolution, I understand that you did not map CMAQ for this figure onto the DP-GC grid.
We now show CMAQ results after applying the DOMINO averaging kernel. Previously, we showed results without the averaging kernel because it was not available for the NASA standard product. Accordingly, we have removed the comparison with that product because the averaging kernel is essential to a rigorous comparison. We also use an overlap function to map the DOMINO pixels onto a 0.5°x0.5° grid. The weighting given to pixels that overlap a given grid box varies with cross-track location and geometric cloud fraction (see Celarier and Retscher, 2009). We also determine the index of the CMAQ grid box associated with each retrieval and use that information and the overlap algorithm to map the CMAQ fields onto the same domain. These steps ensure that the CMAQ output is on the same grid as the satellite-retrieved products.

Figure 5: why do you use the DOMINO product for this figure and not the other products?
It contained an averaging kernel. We now also show the DP-GC product in the new Figure 5.

P17714, L3 "Clearly, care must be taken when drawing conclusions with respect to biases between modeled and satellite-retrieved columns". I think you should remove this sentence and rigorously compare model and OMI products (please see comment on figure 4 and major comments).
Yes, that was a poor choice of words and will be removed. We now use an averaging kernel where appropriate and do not compare with the NASA standard product, which does not contain an averaging kernel.

Section 3.2:
P17716 l 23 I think it is of importance that you better understand the overestimation of the ozone in the model. Figure 9 shows that by adding NO from lightning you scale up the ozone in your simulations and increase the bias between the model and the observations.
We have spent considerable time diagnosing the causes of this bias. We no longer believe it is mostly caused by biases in the boundary conditions. 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 to eliminate or at least lessen this problem. An updated algorithm is expected to be available 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.

Because of these problems we will de-emphasize our analysis of upper tropospheric ozone in this paper. Specifically, we will remove the comparisons with OMI column time series and TES data. We will still show the sonde comparisons and use them to illustrate the problem and discuss the planned solution.

P17718 L5 can you explain, why you think you can use fixed boundary conditions?
We used fixed boundary conditions in 2004 to be consistent with Napelenok et al. (2008). Their results were one of the motivating factors for this project. We don't believe the use of fixed boundary conditions have a substantial impact on our conclusions with respect to the INTEX-A / model comparisons.

Section 3.5: This section provides interesting results. But, it would be interesting to know how the HOx in CMAQ compares with HOx measured during the INTEX-A campaign.
We have added the following paragraph to the text:

When averaged over all INTEX-A flight days, biases in mid- and upper-troposphere HO_x (after multiplying measured HO_x by 1.64 to account by interferences discussed in Ren et al. [2008]) are minor for simulation LNO_x (1.5% too high at 7-9 km and 4.7% too high at 9-12 km) [not shown]. Biases for HO_2 are also small in these altitude ranges (2% high at 7-9 km and 7% high at 9-12 km). Model OH has a low-bias of 21% for 7-9 km and 47% for 9-12 km. Thus the low-bias in NO_x is not believed to be caused by excessive OH.

Overall, model HO_x decreases less rapidly with altitude than observed HO_x resulting in small biases in the mid- and upper-troposphere and larger biases in the lower troposphere. Model HO_x is too low in the boundary layer with low-biases of 20-30% for HO_2 and HO_x and 10-25% for OH.
Figures 2a-b of this response to reviewers show mean plots for HO_x and OH:

Some of the references in the text are missing in the list.
We have double checked the references.

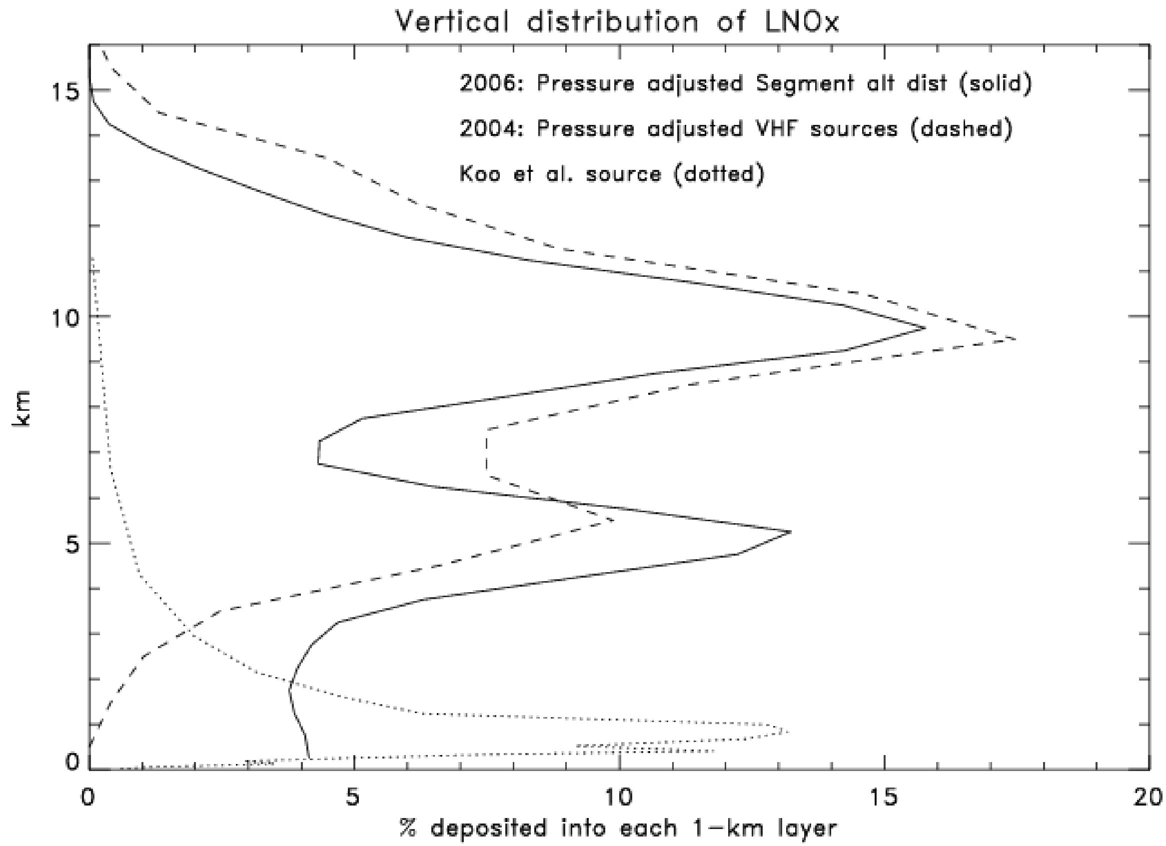

Figure 1. Vertical distribution of lightning-NO production assumed for CMAQ simulations by Allen et al. (2011) and Koo et al. (2010). The solid (dashed) black line shows the Allen et al. distribution for 2006 (2004). The dotted line shows the Koo et al. distribution.

(a) (b)

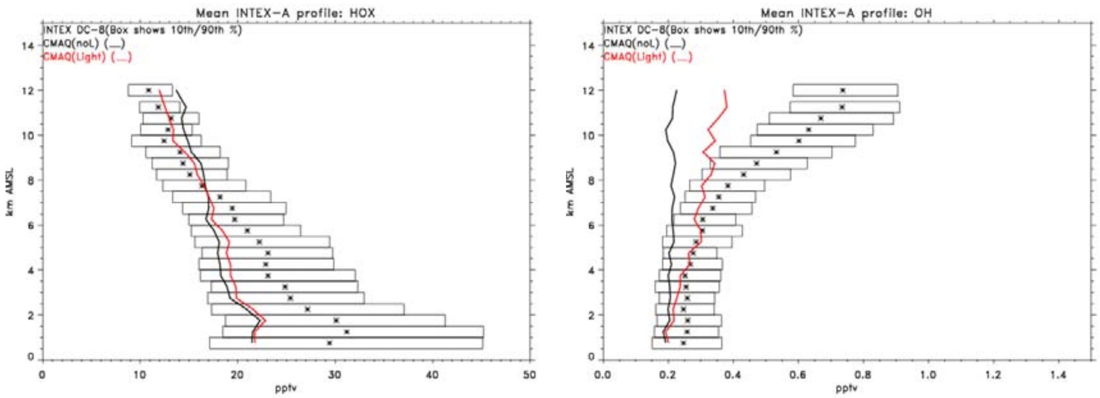

Figure 2. Vertical distribution of HO_x (a) and OH (b) during INTEX-A. Means of medians from 16 DC-8 flights are shown by asterisks. Measured values have been multiplied by 1.64 to account for interferences discussed in Ren et al. (2008). Box edges show mean 10th and 90th percentiles for the 16 flights. Model means of medians from simulations noL (solid black line) and LNO_x(dashed red line) are also shown.