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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of lightning-NO on Eastern United States photochemistry during the summer of 2006 as determined using the CMAQ model" *by* D. J. Allen et al.

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

Atmospheric Chemistry and Physics Discussions General comments:

Anonymous Referee #2 Received and published: 18 August 2011

The manuscript by Allen et al. is potentially suited for publication in ACP, but requires major revisions as explained in detail below.

Major concerns 1. The paper (title, topic, results and figures) is in large parts similar to Allen et al., JGR, 2010. The authors have to clearly announce this in the introduction

and have to clarify what is new/different in this study and in how far a separate publication is justified. We believe there is easily enough new material to justify a paper. In this paper we discuss the causes of differences in NO2 columns between urban and rural sites, the impact of lightning-NO on deposition of nitrogen species, and the sensitivity of upper tropospheric NOx 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 authors announce a "new lightning-NO parameterization" (17703/15). This new scheme has to be set in relation to existing lightning parameterization schemes (see e.g. Tost et al., "Lightning and convection parameterizations", ACP, 2007.) This statement has been removed it from the text. As the reviewer notes, this parameterization is closely related to the parameterization developed in Allen et al. (2010).

The authors have to show the (superior?) performance of their new parameterization scheme, e.g. by adding modeled flash rate distributions from other schemes to Fig. 2 exemplarily. The focus of this paper is not the development of a new lightning parameterization. This paper focuses on the impact of lightning-NO emissions on eastern U.S. photochemistry. We choose to use convective precipitation as a predictor of flash rate because it has been shown to be an indicator of lightning, and because it is routinely archived. In order to test other schemes, we would have to re-run the meteorological simulation archiving additional convective parameters. While this might be interesting, it is beyond the scope of this paper. Accordingly, in the manuscript, we have removed the emphasis on the lightning parameterization, and directed the focus toward evaluating the impacts of lightning-NO on photochemistry and N deposition. .

3. Our knowledge on lightning NOx is still highly uncertain and inconsistent (compare e.g. Beirle et al., ACP, 2010, which is not compatible to a LNO production of 500 moles/flash). The authors indeed mention some discrepancies and shortcomings in the text, but from reading abstract and conclusions only, one might get the impression that by just adding the LNOx to the model, everything works out fine But this is not

the case: We now mention that the conclusions are based on a model and assume a relatively high and uniform source of 500 moles per flash. We will continue to note that a "considerable NO2 low-bias remains in the uppermost troposphere".

a) The fact that upper tropospheric NOx is underestimated by CMAQ might indeed be due to missing LNOx (of course, by adding an upper tropospheric source, a bias can be reduced). But the bias might as well be caused by a wrong/insufficient implementation of chemistry and/or deep convection in the model; the latter should also be discussed by the authors. It is evident from Fig. 17 that there is something completely wrong in the upper troposphere, which can not be fixed by just tuning the LNO source. Yes, the agreement is poor but it is improved after we add lightning-NO emissions to the model. We spend most of section 3.5 examining factors other than missing LNOx that could explain the poor agreement. For example, we discuss the sensitivity of this bias to known uncertainties in tropospheric chemistry and to interferences in NOx measurements. We will modify the text to make it clear that we do not believe missing LNOx is the sole cause of this discrepancy. We have also added the following: Several factors may contribute to the sizeable bias between modeled and measured NOx including biases in model convection, measurements, and model chemistry. For example, if model clouds do not extend high enough into the upper troposphere, the lofting of boundary layer ozone precursors and the vertical extent of NO with a lightning source will be underestimated.

b) The NO2 comparison done by the authors (section 3.1) is not conclusive: The main reason the results were not conclusive is that uncertainties in the satellite-retrieved products are too large. We no longer use the AVDC product and use an updated DOMINO product. Agreement with the model fields is improved, and the improvement in model NO2 due to including lightning-NO is substantial.

b1) Three OMI NO2 products are considered, but only the results for DOMINO are given as numbers in the text; We list the mean, standard deviation, and normalized standard deviation for each of the products. When we compare with CMAQ we focus on

the DOMINO product because it includes averaging kernels making quantitative comparisons with CMAQ more rigorous. In addition to the comparison with the DOMINO product, we have added an additional panel to figure 5, which shows the agreement with the DP-GC product.

for DP-GC, I assume that the addition of LNO even leads to worse agreement, if AKs are considered! Figure 5d now shows model/DP-GC biases after application of the DOMINO averaging kernel to the model fields. Overall, the model column is biased high by $\sim$13%. Yes, this high-bias is increased by averaging kernel processing; however, the increase is not as large as we initially thought. After reprocessing the level 2 DOMINO fields and associated model output using the mapping algorithm of Celarier and Retscher (2008), we find that averaging kernel processing increases the mean column by approximately 8%. We gave a value of 14% in the previous version of the paper.

Numbers for all products have to be given in the text, and the CMAQ avgK columns should be also added to Fig. 4 for better comparison. In version 1 of this paper, we listed the mean, standard deviation, and normalized standard deviation for each of the products. When we compared with CMAQ we focused on the DOMINO product because it included averaging kernels making quantitative comparisons with CMAQ more rigorous. We no longer show the AVDC product as it is based on the NASA standard product which will soon be replaced and is known to have a large high-bias. We have added a more quantitative comparison with the DP-GC product. Also, model fields are smoothed with an averaging kernel before being used in Figure 4.

b2) All number are based on temporal and spatial averages over large scales. The mean LNO2 contribution (0.31e15 molec/cm2) is of the same order of magnitude as the uncertainty of the stratospheric estimation (0.15-0.2e15, Boersma et al., 2007), which is potentially systematic, i.e. is not eliminated by the temporal averaging. Consequently, the observed difference of model and satellite must not be over-interpreted The mean contribution (0.31e15 molec/cm2 for version 1 of paper) was obtained by differencing CMAQ simulations with and without lightning-NO emissions. No averaging kernel was applied and no satellite data were used. We will emphasize that this is a model result.

Given the likely contribution of uncertainties in the stratospheric estimation to these differences, we will give less emphasis to conclusions based on model/satellite differences.

Also, as you note the spatial and temporal scales of the averaging are large. We chose large spatial scales to minimize differences in mean column due to differences in sampling location.

(compare 17714/4-5). b3) The authors admit that the addition of LNOx does not improve the correlation of daily mean NO2 between model and OMI, despite the large-scale averaging. This is of course rather disappointing. It would be quite interesting to investigate if there is at least a correlation of the observed daily NLDN flash rates (prior to the OMI measurement) and the observed OMI columns? If this is not the case, there is no indication at all that OMI columns are affected by lightning, and quantitative conclusions are meaningless. Over the 120°-70°W, 25°-50° N region, the correlation between observed daily NLDN flash rates and observed OMI columns is also quite poor. We believe this is because lightning-NO emissions are responsible for only ~25% of the column. We do not believe this result makes the conclusions meaningless. As part of a separate project, we are identifying time periods and locations where lightning-NO emissions do have a large impact on the total column.

Thus, the authors have to - clearly admit the still existing uncertainties w.r.t LNOx, also in abstract and conclusions, We now give a value for the lightning-NO source both within the abstract and conclusion.

- discuss the a-priori choice of 500 moles/flash (it is at the upper end of the estimates given in Schumann and Huntrieser, but still can not fix the bias in the UT! Yes, LNOx alone cannot fix the upper troposphere bias. We note that in the manuscript. We have

added the following to the text: This value (500 moles per flash) is on the higher end of estimates in Schumann and Huntriesser (2007) and is much higher than a recent top-down estimate obtained by Beirle et al. (2010) from their comparison of observed flash rates and NO2 columns from SCIAMACHY.

In addition, it was found by Jourdain et al., 2009, by comparing model to TES O3 data, while, in this study, the authors clearly state that "LNO algorithms should not be evaluated by how much they improve biases between modeled and measured UT O3", 17718/8-10), Jourdain et al. (2009) is not the primary source of the 500 mole per flash value. It is an average value primarily based on cloud-scale modeling of observed events during the STEREO, CRYSTAL-FACE, and EULINOX campaigns (see Ott et al., 2010).

- clarify, how far the comparison in 3.1. actually tells us anything on LNOx. Section 3.1 is being re-written. Comparisons are now being made with improved satellite products allowing for more robust conclusions. It shows the mean model contribution to the column and also shows the mean model bias with respect to satellite-retrieved columns.

Further comments: The discussion of rural vs. urban regions is interesting in itself, but somehow off topic within this study. I recommend to shorten this discussion in 3.1, and especially in the abstract (17701/14-21).

Another reviewer suggested that I highlight this section. I have modified this section to emphasize its relevance to the air quality community. The paragraph follows: Huijnen et al. (2010) compared tropospheric NO2 columns over Europe from ten different regional models and two global models to the DOMINO product, version 1.0.2. They found that median model columns were too low at rural locations and too high at urban locations. Castellanos et al. (2011) also found high biases at urban locations and local biases at rural locations when comparing compared CMAQ-calculated NOy-HNO3 with "NO2" measurements at rural and urban monitoring sites over the eastern United States. These differing biases are important because they suggest that the lifetime of NO_2 (see Henderson et al., 2011) and/or the transport of NOx (see Gilliland et al., 2008) is underestimated by regional models. These underestimations could lead to errors in inverse-based emissions of sources and to misleading results as to the relative importance of local versus regional emissions.

17701/8: Add "Assuming a LNO production of 500 moles/flash, . . .". Yes, I do need to make the production rate clear, especially since I choose a fairly high one.

17704/10: The DOMINO product is currently updated, see Boersma et al., AMTD, 2011. Please check how far the changes affect your conclusions. I now use v2 of the DOMINO product. When averaged over the region shown in Figure 4(110°-70°W, 25°-50N°) it is 10% lower than v1. Its use reduces biases between the model and DOMINO.

17704/21: When investigating lightning NOx, the selection of cloud free pixels probably introduces a systematic bias, as lightning is generally accompanied by clouds. Please comment on that. Yes, this could be a problem, especially with a weighting scheme such as Celarier and Retscher (2008) that gives more weight to cloud-free pixels. This effect warrants more attention. I've added this to the text: The mean value in each grid box was then obtained by weighting the high-quality retrievals using the algorithm of Celarier and Retscher (2009). This algorithm gives more weighting to pixels with near-nadir field of views than far-off-nadir fields of view and to clear pixels than partly cloudy pixels. Pixels with cloud geometric fractions exceeding 0.3 are given a weighting of 0. Since lightning-NO emissions are associated with clouds, this cloud-dependent weighting could lead to a low-bias in satellite-retrieved columns. In order to minimize the impact of this effect on conclusions, CMAQ profiles are weighted in the same manner as DOMINO profiles

17706/28: Add a reference to Kain-Fritsch parameterization. Done

17713/18: Replace the ";" by a ",". OK.

17714/19: How large would these biases be? Domain-averaged biases are also shown in Figure 4. I now include a panel showing the bias with respect to the DP-GC column.

17715/11: Replace "belief" and give a reference. OK. Will reference Henderson et al.

The text now states "the upper tropospheric lifetime of NOx was determined to be too short in atmospheric models (Henderson et al., 2011)."

17725/8: Add "Assuming a LNO production of 500 moles/flash, . . .". Done.

Figures: The authors should improve the choice of colors in their figures; avoid having the same color+line style for two different data sets as in Fig. 4. The purple in Fig. 17 is hard to recognize. I now use both color and line style to differentiate between the data sets.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C10678/2011/acpd-11-C10678-2011-supplement.pdf

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