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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of convective transport and lightning NO_x production over North America: dependence on cumulus parameterizations" *by* C. Zhao et al.

C. Zhao et al.

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Reviewer 2

General Comments:

(Q): The information within this paper is original and somewhat useful to the regional and global photochemical modeling communities within the ACP readership. However, there are some clarifications and details (see major comments below) that should be addressed before publication.

(A): We thank the reviewer for a detailed review. Both text and figures are revised.

Technical Comments:

(Q): Page 2292, paragraph, lines 11-26. a) A major premise in this article is that the convective parameterization, and not the differences within the WRF and MM5 formulations, is responsible for chemistry differences in the mid and upper troposphere. A clean comparison of convective parameterizations requires the same computational core. Your first sentence of this paragraph states how sensitive convective parameterizations are to the underlying meteorology. The last sentence tells the reader to ignore that fact, and trust your judgment, that convective parameterizations are the responsible for any differences.

(A): We agree. Please see the response to common comments.

(Q): b) The Grell et al. (2002) reference is the convective parameterization currently used within the WRF model. The Grell convection scheme used within MM5 (Choi et al., 2008a) is from a much older reference (1993). There are major differences between the two. Was the MM5 convection scheme updated to the newer reference? My suspicion is that the 1993 Grell scheme was used in this study, which makes this whole comparison rather useless to the community.

(A): We used MM5 v3.6.1, which used Grell convection scheme (1993). We checked that the latest MM5 (v3.7) still used Grell scheme (1993).

The review is correct that Grell scheme in WRF is very different from that in MM5. We now make it clear in the paper.

(Q): c) The reasoning behind using MM5/Grell versus WRF/Grell is the absence of a shallow convection scheme within WRF/Grell. There is no information or references pointing to the importance of shallow convection to regional scale meteorology or transport. I would submit that changes in the dynamic core (MM5 versus WRF) are much more important to convective transport than inclusion or absence of a shallow convection scheme.

(A): We agree. Please see the response to common comments.

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(Q): Page 2293, lines 18-20 Please state the variables that are saved every 5 minutes, and why those particular variables are given higher temporal resolution. If someone were to redo your methodology they should know more details. One important issue that is missing from the paper is the distinction and relative importance between resolved transport and parameterized convection. The temporal resolution of meteorological fields has an impact on this partitioning. Some justification needs to be given for your methodology, especially since there is a publicly available on-line version of WRF (WRF/Chem) that circumvents these issues associated with off-line models.

(A): Only convection related variables are saved every 5 minutes, such as cloud top height, cloud base height, mass fluxes, and convection available potential energy. We now give the list of variables that we save at the higher frequency in the text.

(Q): Page 2293, line 24 to page 2294, line3. The SMOKE emissions processor comes with 3 photochemistry options; SAPRC-99, CBM-IV or CBM-V. Which option did you choose and how did it interface with REAM chemistry? Why didnt you also use ethane and propane emissions from this inventory, since you state that GEOS-CHEM default emissions are inconsistent with INTEX-NA?

(A): We use SPARC-99 option in SMOKE. We partitioned the lumped tracers from SMOKE to the species using the reported species ratios in the emission inventory. We only found >C4 alkanes in GEOS-CHEM is inconsistent with INTEX-NA measurements. The ratio of the emissions of ethane and propane from GEOS-CHEM give a more reasonable comparison with INTEX-A measurements than that from SMOKE. The ethane/propane emission ratio was based on the study by Wang et al. [1998].

(Q): Page 2294, lines 19-24 There are some major differences between the treatment of lightning in this paper, and previous work that is not mentioned. The Cooper et al. (2006) paper uses an IC/CG ratio of 4.2, and 457 mole/flash of NOx. How were the 250 moles NO/flash determined? This needs some clarification, and reference to previous lightning and NOx source estimates.

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(A): We set a NOx production rate of 250 moles NO per flash in this study because we found that model simulations with this rate are more consistent with in situ and satellite observations (through trial and error). And also, the rate is consistent with that suggested by Schumann and Huntrieser [2007]. It is now clarified in the paper.

(Q): Page 2295, lines 5-17 VOC measurements are from canister samples. Is there interpolation to put them on the 1-minute merged dataset?

(A): We now use the original VOC canister sampling measurements for ethane and propane comparison. The result is similar.

(Q): Page 2297, line 24 to page 2298, line 2, C2H6/C3H8 discussion a) State specifically which flights in the Singh et al. (2006) reference are used in this data. Also, Figure 3 should have some indication of the number of samples that went into the 1km bin determination of the observations.

(A): The data collected in all the flights over the North America outflow regions (over the western North Atlantic). The number of data points is mentioned in the paper.

(Q): b) Are the model results shown in Figure 3 for the times and locations of the aircraft sampling, or are these medians from the entire 2 month run over a pre-defined region?

(A): Yes. When we show the comparisons between the measurements and simulations, all the model outputs are sampled at the time and location of in situ measurement. It is now clarified in the paper.

(Q): c) Using C2H6/C3H8 ratios, rather than absolute C3H8 or C2H6 concentration comparisons with observations can hide model deficiencies and introduce compensating errors. One critical quantity is the background C2H6 concentration supplied upwind from the GEOS-Chem model. Another critical quantity is the emission ratio of C2H6/C3H8 over the U.S. in your simulations. Maybe background C2H6 is high by 50 percent, the models would match observations without convection, and WRF/REAM has too much convective transport. Though it is reassuring to see that both models are

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somewhat consistent when convection is turned off, comparisons of absolute values of C3H8 with the observations would provide needed credibility to your case, and give skeptics like myself some confidence that the ratios are meaningful, and are not just being used in order to mask some underlying inconsistencies.

(A): We include ethane and propane comparison in the Appendix (see also the response to common comments).

(Q): Page 2299, lines 3-19, HNO3 discussion Without supporting figures this discussion is hard to follow. Moreover, HNO3 will depend on the assumed NOx production rates from lightning. Since these are uncertain within a factor of 2, and the rain-out treatment is so crude and model-dependent, it seems this paragraph has little to offer in terms of useful information.

(A): We include HNO3 comparison in the Appendix (see also the response to common comments). The lifetime of NOx is not assumed; it is calculated online.

(Q): Page 2299, lines 18 to 26, page 2300 a) I am familiar with another SCHIAMACHY comparison over the eastern U.S. between June and August of 2004 (Kim et al., GRL, VOL. 33, L22812, doi:10.1029/2006GL027749, 2006). In that study the authors used a more restrictive criteria (cloud fraction < 15 percent) and found much higher occurrences of SCHIAMACHY data available for use (a minimum of 4 and a maximum of 10 samples) for any given location over the 3 month period. This number of 2 for a 2 month period seems too low, particularly since the criteria is looser.

(A): The results are consistent. We had a minimum of 2 measurements from SCIAMACHY for 1.5 month during INTEX-A (July 1st-August 15th) which is consistent with the previous study [Kim et al., 2006], i.e., 4 measurements for 3 months. The cloud fraction of <15 percent has almost the same effect with that of <30 percent in data filtering. The study period is clarified in the paper.

(Q): b)The comparisons in figure 4 raises some obvious questions: what would Figure

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4 look like with 457 mole NO/flash produced instead of 250?

(A): NOx production per flash, IC/CG flash ratio, IC/CG NOx production ratio, and NOx lifetime (if assumed; not apply to this study) are all variables affecting the resulting NOx distribution. For our setup, we chose 250 moles per flash on the basis of observations and previous studies. There are no enough observations to independently and conclusively determine all the parameters related to lightning NOx production.

(Q): How accurate were cloud tops predicted by the 2 models on the 2 days with cloud free observations?

(A): The point of the comparison is partly that SCIAMACHY type of measurements at the moment does not provide enough quantitative constraints to differentiate the model results.

(Q): Both models use the same vertical profile for NOx injection, but wouldn't discrepancies be even larger if IC flashes were apportioned vertically according to cloud top?

(A): NOx from IC flashes are apportioned vertically according to cloud top height. Please see Pickering et al. [1998] for the dynamic reasons of not using separate profiles for IC and CG lightning flashes.

(Q): Page 2301, lines 2 and 3, reference to Figure 5: a)The values of lighning NOx near the surface should probably be mentioned. These appear significant, and should affect rural surface level O3 values. Is this due to the Pickering (1998) profile assumed? Is MM5 NOx at the surface so much higher because of the NOx emission parameterization, or is it because convection is bringing lightning NOx down to the surface more efficiently? b) Only a suggestion: If surface O3 is affected significantly, this would also have implications for previous lightning NOx assessments on surface O3 (Kaynak et. al., Atmos. Chem. Phys., 8, 5151-5159, 2008). It may be useful to at least qualitative state if your results (at the surface) are consistent with that reference.

(A): We estimate the vertical distribution of lightning NOx following Pickering et al.

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[1998] (stated in the paper). The distribution is different from Kaynak et al. [2008]. It is only one of many areas that the model setup in this work differs from that paper. We did not look at the impact of the lightning NOx on surface O3 in this study. The focus of this study is in the upper troposphere. A detail comparison to Kaynak et al. [2008] would require a separate paper. Since our lightning is consistent with model convection (not the case in the work by Kaynak et al. [2008]), neither cloud effects on radiation nor fast transport associated with convective systems favors O3 production. The model details would be different but the impacts of lightning NOx in our model should be just as small. We avoid model-model comparisons when there is no observational constraint.

Lightning NOx from MM5-REAM at the surface is higher because of its larger total lightning NOx production (from intra-cloud lightning).

(Q): Page 2301, line 25 to top of page 2302: a)The presentation within figure 7 used to make the relationship between increased NOx and increased O3 obscures or ignores a lot of useful information. While the first 3 panels (left to right) are appropriate and useful, the last panel on the right (differences) is hard to digest, and is only used to say that 20 ppbv of additional O3 is possible. It seems more useful to show NOy and O3 differences over the entire domain (averaged over the 2 months) or vertical distributions of O3 changes averaged over the same area as in Figure 5.

(A): The comparison is meant to demonstrate the effects of lightning NOx on O3. So the last column is important to include. The effect of lightning NOx may be apparent from the previous three columns; the effect on O3 is not. The figure is used to illustrate in situ observations cannot be used to differentiate between the two model results. Time averaged comparison does not serve that purpose.

(Q): Page 2302, lines 19-20: Please be more specific about the area used to derive these percentages. Is this over the model domain, eastern U.S. or what?

(A): It is over the INTEX-A region. We now clarified in the paper.

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(Q): Page 2302, lines 20-28, page 2303 lines 1-7: A simple figure would help the reader digest this information more easily. It is somewhat ambiguous, line 21 says up to 70 percent lightning NOx contribution in the 8-12km region. But page 2303 lines 3-4 say 40 percent.

(A): We stated "Lightning contribution to NO increases from 10 percent in the boundary layer to up to 70 percent in the upper troposphere (8-12km)." and "We also estimate contributions of 40 and 10 percent to total reactive nitrogen (NOy) from lightning and surface emissions at 8-12km, respectively." The distinction is quite clear. One would expect a lower lightning impact on NOy because of long-range transport.

(Q): Pages 2303-2304, Conclusions: You cannot say that 2 convective schemes were compared. Output from two different models, using two different convective schemes were compared.

(A): We agree. Please see the response to common comments.

Minor comments:

(Q): Page 2292, line 5, GOES-CHEM model should be GEOS-CHEM; Page 2292 line 11, Grammar, sensitive with should be sensitive to; Page 2292 line 16, Grell et al. (2002) should be Grell and Dévényi (2002); Page 2294 line 1, spell out VISTAS acronym.; Page 2294, lines 8-12, and Figure 1b Do the fluxes for the KF-eta model include the detrainment/entrainment fluxes shown in the plot? This should be stated.; Page 2297, lines 28, also Page 2298, line 9: no reference to Wang and Zeng (2004) in reference list.; Page 2316, Figure 6. What are the holes in the 300-400 hPa cloudtop interval? Some explanation is needed.

(A): All the above are corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 2289, 2009.