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Interactive Comment

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

O. Cooper (Editor)

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This review is by Owen Cooper, co-editor of ACPD/ACP and the editor of this manuscript.

Analysis of cumulus parameterizations and quantification of their impact on trace gas distributions is an important topic and entirely relevant to ACP. Overall I find the paper to be well conceived and written but it has two major deficiencies that need to be addressed before publication: 1) utilization of the DC8 NO data and 2) a more extensive comparison to previous work. I have read the comments of the first referee and am in agreement with his/her assessment. However, I have not yet had the benefit of reading the comments of the second referee which will have a strong impact on my final decision regarding the publication of this manuscript.

1) When simulating the production of lightning NOx it is ideal to verify the model against measurements of NO+NO2 as 80-90% of upper tropospheric NOx is in the form of NO. In this analysis you have chosen not to use the DC-8 NO data for the following reason: "The NO measurements on the DC-8 are not used due to its limited sensitivity only suitable for measuring mixing ratios larger than 100 pptv [Singh et al., 2007]." I spoke to Dr. Xinrong Ren who made the NO measurements. According to Ren et al. [2008]: "measurements of NO were made with a TECO Model 42C NO-NOx analyzer run in an NO only mode, which had a precision of 50 pptv with 1-min time integration."

So basically below 100 pptv the NO data are noisy, but not biased. The instrument noise would be a problem if you were trying to model ozone production for a specific time and location in a low NOx environment. But for the purpose of your study you are interested in comparing model output to measured values averaged over a fairly large area, in which case the noise from the instrument is averaged out. Also, you are interested in the upper troposphere which has few values below 100 pptv. The DC8 NO+NO2 data have been used in previous studies to verify LNOx production [Cooper et al., 2006, Hudman et al, 2007], and they are fine for these purposes when averaged over a fairly large domain. In my conversation with Dr. Ren he said that if you wanted to be as accurate as possible you can calculate NO from photostationary state using measurements from the DC8 and the equation: [NO_pss]=J(NO2)*[NO2]/(k_O3_NO*[O3]+k_HO2_NO*[HO2]+k_CH3O2_NO*[CH3O2_model+RO2_model])

Modeled HO2 and RO2 are needed and they are available in the INTEX-A data archive (LaRC box model results) and the same rate constant is assumed for the reactions of CH3O2+NO and RO2+NO (here RO2 excludes CH3O2). Because typically {k_O3_NO*[O3]} is much greater than {k_HO2_NO*HO2+k_CH3O2_NO*[CH3O2_model+RO2_model]}, the above equation can be simplified as:

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NO_pss=J(NO2)*[NO2]/(k_O3_NO*[O3])

Alternatively, Dr. Ren said he would be willing to share the calculated NO values with you (xren@rsmas.miami.edu). In your paper please focus on NOx rather than NO2. This will make the results more relevant to those interested in lightning NOx production and will also facilitate comparison between your results and those of other researchers.

Along these lines, measured NOx needs to be included in Figure 5. Please include standard deviations of the measured and modeled values.

2) Over the past few years quite a few studies have appeared that discuss the importance of lightning NOx on the summertime production of upper tropospheric ozone [Cooper et al 2006, 2007 and 2009; Hudman et al 2007 and 2009; Li et al 2005; Pfister et al 2008, Zhang et al. 2003]. Those studies are mentioned throughout this review and the references are listed below. It is important for you to place your results in the context of these earlier studies. Please read through these papers and in the Discussion section of your paper compare their results to yours, focusing on the following. a) How well do the various studies model upper tropospheric NOx (magnitude and spatial location) in comparison to the observations? b) Do they all agree that LNOx is the most important factor for upper tropospheric ozone production above N. America during summer?

Other comments: Are the PIs who made the C3H8, C2H6, HNO3, NO2, and O3 data aware that their data are being used in this paper? I ask because I do not see their names in the author list or the acknowledgements. The data may now be freely available but the PIs should still be given the opportunity to be co-authors, or at the very least, be acknowledged at the end of the paper. They will also be able to give feedback if their data have any known problems. Acknowledgement should also be given to the ISCCP team. Please check their website to see if they have a standard acknowledgement statement.

Figure 6 Over some regions of the Gulf of Mexico and nearby Atlantic, the MM5 simulation seems more accurate. Ridley et al. [2004] report NOx measurements during CRYSTAL-FACE over Florida and show that LNOx is found at higher altitudes (14 km) than above mid-latitude USA. So the Grell scheme may be more accurate for subtropical/tropical conditions.

Page 2301 More care needs to be taken when summarizing the ozone production due to LNOx in earlier studies. The values of 10 ppbv from Hudman et al [2007] and 11-13 ppbv from Cooper et al [2006] are just the average values for the eastern USA. Ozone production could be much more in other locations. For example, Cooper et al. [2006] report that ozone production from LNOx could account for the full 24 ppbv ozone enhancement above Texas during summer 2004. Also, Cooper et al. [2007] report simulations by an atmospheric chemistry general circulation model that indicate lightning NOx emissions led to the production of 25-30 ppbv of ozone at 250 hPa above the southern United States during August 2006. Other papers that estimate ozone production from lightning above the USA and that need to be mentioned in your paper are: Hudman et al 2009; Pfister et al., 2008 and Zhang et al 2003.

Minor comments: If no reason is given for a comment then it means that the suggested text should replace the corresponding text in the manuscript.

page 2290 line 10 NOx production.

page 2291 line 8 western North Atlantic Ocean

page 2291 line 19 Hudman

page 2292 line 5 GEOS

page 2292 line 6 What is the reference for 1-20 Tg N/yr? A more recent estimate (2-8 Tg N/yr) can be found in the excellent review of LNOx production by Schumann and Huntrieser, 2007.

page 2292 line 11 Modeled convective transport of tracers and lightning NOx production are sensitive to underlying.....

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page 2292 line 15 based on the KF-eta [Kain et al., 2003] and the Grell [Grell et al., 2002] convection schemes

page 2294 line 23 Who determined from INTEX-NA data that the NOx production rate should be 250 moles NO/flash? Please provide the reference. By the way, this agrees with the mid-range value discussed by Schumann and Huntrieser [2007].

page 2295 line 20 The SCHIAMACHY instrument onboard the ENVISAT satellite has a

page 2296 line 7 cloud climatology based on the images

page 2297 line 5-7 How do the updrafts and entrainment/detrainment in WRF scavenge more soluble species? More contact time between the gases and droplets? Allowing more air to flow through the storm?

page 2297 line 10 3.2 Convective impact on export of pollutants

page 2297 line 11 This sentence needs to be rewritten as it implies that soluble species are not lofted. Convective transport lifts both soluble and non-soluble pollutants from the boundary layer to the free troposphere. Whether or not the soluble species remain in the free troposphere depends upon precipitation.

page 2297 line 15 driven

page 2298 line 4 Please define the domain of the western North Atlantic outflow region. Is it just over the water or does it also include the eastern USA. If it does not include the eastern USA, then why leave this region out which was heavily sampled by the DC8 and which also experienced a lot of convection?

page 2298 When comparing the model to the DC8 data, did you sample the model just at the times and locations of the DC8 flight tracks or is it a regional average?

page 2299 line 5-9 It's not clear if you are talking about the models or the real world.

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page 2299 line 12 Seeing as a unique aspect of this paper is its verification against in situ measurements, please show the HNO3 data in a figure.

page 2299 line 22 What do you mean by 2 measurements? Two measurements per pixel over the entire 62 day study period?

page 2300 line 3 What do you mean by lower? Lower altitude or lower in quantity?

page 2300 line 13 Which model are you referring to, just WRF-REAM?

page 2300 line 21 cloud-to-ground

page 2301 line 18 indicate that the model

page 2302 line 14 The reference to Grewe et al [2001] regarding the lightning contribution to upper tropospheric NOx is somewhat outdated compared to the more recent studies listed below. Please also mention the results of the later studies.

page 2302 line 28 than those from

Papers mentioned above:

Cooper, O. R., et al. (2006), Large upper tropospheric ozone enhancements above midlatitude North America during summer: In situ evidence from the IONS and MOZAIC ozone measurement network, J. Geophys. Res., 111, D24S05, doi:10.1029/2006JD007306.

Cooper, O. R., et al. (2007), Evidence for a recurring eastern North America upper tropospheric ozone maximum during summer, J. Geophys. Res., 112, D23304, doi:10.1029/2007JD008710.

Cooper, O. R., et al. (2009), Summertime buildup and decay of lightning NOx and aged thunderstorm outflow above North America, J. Geophys. Res., 114, D01101, doi:10.1029/2008JD010293.

Hudman, R. C., et al. (2007), Surface and lightning sources of nitrogen oxides over the

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United States: Magnitudes, chemical evolution, and outflow, J. Geophys. Res., 112, D12S05, doi:10.1029/2006JD007912.

Hudman, R., L. T. Murray, D. J. Jacob, S. Turquety, S. Wu, D. B. Millet, M. A. Avery, A. H. Goldstein, and J. Holloway (2009), North American influence on tropospheric ozone and the effects of recent emission reductions: Constraints from ICARTT observations, J. Geophys. Res., doi:10.1029/2008JD010126, in press.

Li, Q., D. J. Jacob, R. Park, Y. Wang, C. L. Heald, R. Hudman, R. M. Yantosca, R. V. Martin, and M. Evans (2005), North American pollution outflow and the trapping of convectively lifted pollution by upper-level anticyclone, J. Geophys. Res., 110, D10301, doi:10.1029/2004JD005039.

Pfister, G. G., L. K. Emmons, P. G. Hess, J.-F. Lamarque, A. M. Thompson, and J. E. Yorks (2008), Analysis of the Summer 2004 ozone budget over the United States using Intercontinental Transport Experiment Ozonesonde Network Study (IONS) observations and Model of Ozone and Related Tracers (MOZART-4) simulations, J. Geophys. Res., 113, D23306, doi:10.1029/2008JD010190.

Ren, X., et al. (2008), HOx chemistry during INTEX-A 2004: Observation, model calculation, and comparison with previous studies, J. Geophys. Res., 113, D05310, doi:10.1029/2007JD009166.

Ridley, B., et al. (2004), Florida thunderstorms: A faucet of reactive nitrogen to the upper troposphere, J. Geophys. Res., 109, D17305, doi:10.1029/2004JD004769.

Schumann, U., and H. Huntrieser (2007), The global lightning-induced nitrogen oxides source, Atmos. Chem. Phys., 7, 3823-3907.

Zhang, R., X. Tie, and D. W. Bond (2003), Impacts of anthropogenic and natural NOx sources over the U.S. on tropospheric chemistry, Proc. Natl. Acad. Sci. U. S. A., 100, 1505-1509, doi:10.1073/pnas.252763799.

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

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