

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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Editor's comments:

General Comment:

(Q): 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.

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

Technical Comment:

(Q): (1) When simulating the production of lightning NO_x it is ideal to verify the model against measurements of NO+NO₂ as 80-90% of upper tropospheric NO_x 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-NO_x 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 NO_x 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 LNO_x 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: $[NO_{pss}] = J(NO_2) * [NO_2] / (k_{O_3-NO} * [O_3] + k_{HO_2-NO} * [HO_2] + k_{CH_3O_2-NO} * [CH_3O_2_model + RO_2_model])$ 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 $\{k_{O_3-NO} * [O_3]\}$ is much greater than

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{ $k_{\text{HO}_2\text{NO}}\text{HO}_2 + k_{\text{CH}_3\text{O}_2\text{NO}}[\text{CH}_3\text{O}_2\text{model} + \text{RO}_2\text{model}]$ }, the above equation can be simplified as: $\text{NO}_{\text{pss}} = J(\text{NO}_2) \cdot [\text{NO}_2] / (k_{\text{O}_3\text{NO}} \cdot [\text{O}_3])$ 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 NO_x rather than NO₂. This will make the results more relevant to those interested in lightning NO_x production and will also facilitate comparison between your results and those of other researchers. Along these lines, measured NO_x needs to be included in Figure 5. Please include standard deviations of the measured and modeled values.

(A): We now reference Ren et al. (2008) and use NO_x in relevant comparisons.

(Q): 2) Over the past few years quite a few studies have appeared that discuss the importance of lightning NO_x on the summer time production of upper tropospheric ozone [Cooper et al 2006, 2007 and 2009; Hudman et al 2007 and 2009; Li et al 2005; Phister 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 NO_x (magnitude and spatial location) in comparison to the observations? b) Do they all agree that LNO_x is the most important factor for upper tropospheric ozone production above N. America during summer?

(A): We updated the comparison between our study and relevant previous studies in the paper.

(Q): 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 coauthors, or at the very least, be acknowledged at the end of the paper. They will also be able to give

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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.

(A): As all the measurements in INTEX-A and from ISCCP used in our study are published, we added citations to relevant references and acknowledgement in the paper.

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

(A): Yes, over Florida, MM5 simulated cloud top height is more consistent with observations than WRF. That may indicate that the entrainment and detrainment are overestimated in WRF with KF-eta over that region. We mentioned it in the paper now.

(Q): Page 2301 More care needs to be taken when summarizing the ozone production due to LNO_x 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 LNO_x 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 NO_x 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.

(A): The simulated O₃ impact in this model is similar to Hudman et al. [2007]. So we will limit our comparison to previous studies on the lightning NO_x impact.

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Minor Comment:

(Q): page 2290 line 10 NO_x 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 LNO_x production by Schumann and Huntrieser, 2007; page 2292 line 11 Modeled convective transport of tracers and lightning NO_x production are sensitive to underlying.....; page 2292 line 15 based on the KF-eta [Kain et al., 2003] and the Grell [Grell et al., 2002] convection schemes;

(A): The above are all corrected.

(Q): page 2294 line 23 Who determined from INTEX-NA data that the NO_x 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].

(A): We set a NO_x 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). It is now clarified in the paper.

(Q): page 2295 line 20 The SCHIAMACHY instrument onboard the ENVISAT satellite has a; page 2296 line 7 cloud climatology based on the images;

(A): The above are all corrected.

(Q): 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?

(A): More updrafts remove more HNO₃ lifted from the boundary layer. Entrainment brings background (out of cloud) HNO₃ into cumulus clouds and HNO₃ is removed. It is mass dependent.

(Q): page 2297 line 10 3.2 Convective impact on export of pollutants

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(A): Corrected.

(Q): 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.

(A): The soluble species are not lofted because they are all scavenged during convection. Much of the convective scavenging occurs as the updraft passes through clouds and soluble species are removed. Below-cloud rain out is usually less.

(Q): page 2297 line 15 driven

(A): Corrected.

(Q): 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?

(A): The outflow region is only for the western North Atlantic Ocean. The reason is to avoid the large standard deviation of the measurements over the polluted regions of the continent.

(Q): 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?

(A): We added that model results are sampled at the times and locations of the measurements.

(Q): page 2299 line 5-9 It is not clear if you are talking about the models or the real world.

(A): It is a discussion of physical processes included in the model. These processes should be present in the real world, although models tend to simplify real-world pro-

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cesses.

(Q): 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.

(A): They are now shown in the Appendix.

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

(A): There were 2-day measurements for the grid over the INTEX-A period (July 1st-August 15th). It is not for 2 months.

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

(A): We clarified that it is lower quantity.

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

(A): We added "in both models".

(Q): page 2300 line 21 cloud-to-ground

(A): Corrected.

(Q): page 2301 line 18 indicate that the model

(A): Corrected.

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

(A): Updated.

(Q): page 2302 line 28 than those from

(A): Corrected.

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