

Interactive comment on “Cloud-scale model intercomparison of chemical constituent transport in deep convection” by M. C. Barth et al.

M. C. Barth et al.

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We appreciate the comments given by the reviewer and thank him/her for his/her interest in our paper. In response to the referee’s general comments, we have included more discussion comparing the different lightning-NO_x parameterizations. Responses to the specific comments of the referee are given here.

Specific comments:

1. *Figures. It would be helpful to add a figure showing the temperature profile measured during the event (the altitude of the initial tropopause level could be seen, as it seems to impact the modelling results of ozone in the upper troposphere) and the initial temperature profile used in the models to trigger convection (warm bubble). This might be done adding a panel in Figure 1.*

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Two panels have been added to Figure 1. The initial water vapor profile and a skew-T diagram of the temperature and dew point temperature are shown.

2. p. 8039 line 12: *please give a typical lifetime for CO and O₃ to show that it is significantly higher than the lifetime of a convective storm.*

The chemical lifetime of CO is typically 1-2 months while that of O₃ is typically 8 days at the surface, 15 days at 5 km elevation, and 40 days at 10 km elevation for summer at 40°N (Brasseur et al., 1999). We have inserted “(days to months)” in the noted sentence.

3. p. 8039, line 19: *please give the acronym for STERAO. Is there a specific reason for choosing this case for the intercomparison?*

The acronym is spelled out now. Further, a sentence has been added stating that the comprehensive set of measurements and the experience of modeling this storm before make this an appropriate storm for an intercomparison.

4. p. 8040: *concentration unit. Volume mixing ratio (which is equivalent to the molar ratio) is more frequently used than molar ratio. It would be better to replace nmol.mol⁻¹ by ppbv, pmol.mol⁻¹ by pptv, etc.*

ACP requests SI units which are what molar mixing ratios are.

5. p. 8040, line 15: *“obtained from the literature”. Please give the reference.*

A citation to Cohan et al., 1999 is now included. Also included is a note that the initial profiles are similar to Snow et al., 2007 although their measurements were taken after the intercomparison exercise was designed.

6. Section 3. *Description of the models. Each subsection describes one of the models used for this intercomparison and is written by the concerned researchers. The information given in each paragraph is not always consistent from each model. For example, the radiative scheme is not always specified. The top boundary condition does not appear for some models (e.g.*

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Wang's model). Please make sure that the same number of information appears for each model (and in the same order). I also noticed that the grid domain is not always the same and the vertical resolution is not the same. I suppose that the domain was chosen in order to get the best modelling result for one specific model. Whatever it is, the reason should appear in the text. Time step: When the time step is specified, please explain if it is the meteorological or the chemical time step. Lightning NOx scheme: it is not always clear to me what is produced by lightning in the models. Is it really NO, NO₂, or a partitioning of these species. Table 2: it should appear more clearly whether the tracking in ice is included or not.

The model description section has been revised to be more consistent from model to model.

7. p. 8041, line 15: "daytime chemistry". What is meant by that? A chemical scheme that includes photolysis reactions?

The referee is correct that "daytime chemistry" means that it includes photolysis reactions, but it also means that species that are normally in small concentration during day (NO₃ and N₂O₅) are ignored. The word "daytime" has been omitted – to reduce any possible confusion, and the species are listed in the next sentence.

8. p. 8053, lines 10-15. Double moment scheme versus single moment: Why RAMS and DHARMA have anvils similar in width to the model with single-moment scheme? More generally, can a general conclusion be drawn in this study about the importance of the type of microphysics on the modelling results? It could help a cloud scale modeller or a future user to go toward one of these schemes.

Expanding the discussion to include detailed explanations of the effects of microphysics on anvil characteristics would not only be beyond the scope of this work but it would detract from the focus on tracer transport. The comparison of storm structure is included for context for the tracer transport comparison. Further, intercomparisons for cloud microphysics (e.g., Grabowski, 2006) and cloud parameterizations (GEWEX) are frequently conducted.

9. Section 4 *Why 3 warm bubbles were used in the simulation while the observations clearly show a double cell structure? Is it because the chemistry results fit better with the observations? It is noticed later in the study (p. 8058 line 13) that all of the models overpredict the air mass flux in the anvil. What would have been this flux if two warm bubbles had been used instead?*

The initiation was determined by the Skamarock et al. 2000 study based on giving the best representation of the storm structure and evolution using the COMMAS cloud model. Further, the COMMAS model (with 3 convective cells at $t=1$ hour) produced air mass fluxes similar to observations. Two to four convective cells were observed during the multicell stage of the storm, thus 3 convective cells at 1 hr of integration is not in error of the observations. Using a two bubble initiation in WRF-AqChem, we find that the storm still transitions from a multicell storm to a quasi-supercell, but is smaller (both horizontally and vertically) with weaker updrafts, and a smaller anvil cross-section. This results in only minor effects on the tracer transport: the magnitude of the anvil enhancement or depletion is similar to the 3 bubble results for CO, O₃, CH₂O, H₂O₂, and HNO₃, the magnitude of NO_x in the anvil is 20-30% greater because of the smaller volume that the NO is placed in, the fluxes of air mass and CO are the same because of normalizing the flux by the anvil area (which is 3 times smaller), and the flux of NO_x is greater.

10. Section 4.2 p. 8054, lines 11-12: *“to a derived cross section obtained from several transects”*. Please add a few words about how this cross section was obtained and above all what is the uncertainty associated with this method.

The methodology is presented a few paragraphs later. We have added 2 sentences regarding the uncertainty related to the objective analysis in this same paragraph.

11. p. 8055, lines 2-8.

a. *In Figure 6, the DHARMA and Spiridonov models depict an increase of NO_x during convection, although the production of NO_x by lightning is not included in the models. Is this due to vertical transport? This should appear in the text.*

The referee is correct. We have added a sentence to make this clear.

b. Lines 6-8: I do not fully agree with this sentence because of the results from RAMS. This model includes a parameterization which is used by a significant number of mesoscale/large scale models. It is surprising to see that for the 10 km downwind panels, the RAMS results are only slightly higher than the DHARMA and Spiridonov models. Could the RAMS user authors comment on this? Would another choice for the transect (slightly shifted in time, in location, or in altitude) lead to the same results? I think a more detailed discussion should be written here about the lightning NO_x parameterization.

While the location and time of the transect play a role in the magnitude of the results shown in Figure 6, the lightning-NO_x parameterizations are also important to the results. One of the key factors in these parameterizations is the source location of the NO produced from lightning. The Pickering et al. (1998) scheme places intracloud lightning-NO_x sources above the -15°C isotherm to cloud top at reflectivity > 20 dBZ (and most parameterizations use this 20 dBZ criterion, but different altitude criteria). An examination of Figure 4 of our paper shows that the 20 dBZ region varies among the model results. The NO_x produced by lightning by the RAMS model is smaller for the IC flashes (111 moles NO/IC flash) but is placed in a relatively large region, resulting in a more reduced NO_x mixing ratio in the anvil. The UMd/GCE model produces more NO/IC flash but has a much larger > 20 dBZ region, also resulting in reduced NO_x mixing ratios at t = 3600 s. In contrast, the explicit parameterizations (Meso-NH and SDSMT) have low NO production per flash but place the NO along the lightning channel – a small volume. This discussion is now included in the paragraph discussing the NO_x transect.

12. From the information provided in the manuscript, it seems that the conclusion is that the Explicit Electrical Scheme (in Meso-NH) is needed for a correct modelling of NO_x at a local scale. Thus I would be less optimistic in the sentence lines 17-19. I would replace "that model parameterizations are capturing..." by "that some of the model parameterizations are capturing..." Please note that the conclusion is not the same at a wider scale since the RAMS model

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does a rather good job in the vertical distribution of NO_x (Fig. 10) and in the fluxes (table 3).

We appreciate the referee's thoughtful comments but suggest that one cannot make such a strong conclusion about lightning- NO_x parameterizations. First of all, several models (with both explicit and parameterized lightning- NO_x schemes) have good agreement with observations locally (Fig. 6). Second, the local and wider results are dependent on several factors (e.g., well represented storm; NO production rate; NO source location relative to the updrafts and downdrafts; similar kinematics to the observations) which could skew the results. Lines 17-19 have been rewritten to indicate the importance of the key parameters to lightning NO_x production but that these parameters also contribute to the uncertainties in NO_x mixing ratios in the convective outflow.

13. p. 8056, 1st paragraph. *Again, is there a further conclusion to be drawn here from the results with single-moment scheme models versus double moment scheme models?*

While it would be nice to provide more information for the cloud microphysics community, the ice particle concentration results are not appropriate for making conclusions regarding single-moment versus double-moment schemes. A much better comparison would be for the mass of ice, which is what the single-moment scheme predicts and would therefore eliminate a significant assumption.

14. p. 8056. *O3 cross section in the upper troposphere. The results obtained by C. Wang and RAMS model are interesting and need further comments. It is stated that the high O3 concentration at the top of the anvil may be due to the strength of the updraft in connection with turbulent mixing at the tropopause. Please remind here the initial altitude of the tropopause level (a dotted line could be added in Figure 9). Would turbulent mixing be efficient at the time scale of the simulation to be seen in Figure 9? It is well known that wave activity (especially when the wave breaks) generated by convection may favour the transport of species across the tropopause. Is a gravity wave activity computed by the models? Which ones? Does it depend on the top boundary conditions? Could the results shown here only be due to a reversible vertical*

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displacement of the tropopause during convection? To check this, the evolution of the isentropic level of the initial tropopause height could be investigated. Another point to be discussed is the potential role of lightning NO_x on ozone formation in the upper troposphere. I expect this process to have a small effect at the time scale of the simulation, but this may be not negligible, especially before sunset. Please check.

While we appreciate the referee's interest in this topic, this paper, which is focused on tracer transport from the boundary layer to the upper troposphere, is not focused on the role of gravity waves enabling cross tropopause transport. These models may or may not capture gravity wave generation or turbulent mixing properly. While the time steps for a few of the models may be appropriate, the horizontal resolution may not (Lane and Knievel, 2005). We have revised the paragraph to de-emphasize reasons for the high O₃ region. Lastly, calculations using the UMd/GCE model results indicate that at most 2 ppbv of O₃ is produced from NO_x generated by lightning. In comparison to 60-100 ppbv, this is small to negligible.

*15. p. 8057. NO_x and NO cross section: You compare NO observations with NO_x model results. In order to make the comparison easier in Figure 10, I propose to plot NO(observed)*1.3. This time, the RAMS model does a better job than for the results shown in Figure 6. The NO_x flux computed by the RAMS model (table 3) is very close to the flux deduced from observations (as for the C. Wang model). On the contrary, Meso-NH computes a lower flux than observed while it is doing a very good job along the transects in Figure 6. Could it be concluded from this that the parameterization of Pickering et al., (1998) is better tailored to regional scale/large scale studies than to very local studies? Is this the contrary for the parameterization of Barthe et al., (2005) within Meso-NH? I think such a discussion would improve the manuscript since one of the aims of this study is to improve the parameterizations related to the transport of chemical species by convection or related processes (Cf. Introduction p. 8038 lines 28-29)*

We think the referee has a very good point regarding further discussion of the lightning-NO_x parameterizations. While the Pickering et al. (1998) scheme is designed for the large-scale or regional-scale model, it cannot be concluded that it does a better job

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than any of the other parameterizations presented here. As pointed out above, the numerous parameters that contribute to lightning- NO_x production are not straightforward and therefore contribute significant uncertainty to NO_x mixing ratios. We have added more discussion about the lightning- NO_x parameterizations after the NO_x flux comparison. The Figure 10 has not been changed for one important reason. The 1.3 scaling factor of the NO_x flux is a mid-value of 1.1 to 1.6 estimated for the actual storm (Skamarock et al., 2003; Dye et al., 2000). Because of this actual variation, showing $1.3 \cdot \text{NO}$ for the observations would be just as ambiguous as showing no scaling factor as is currently done.

16. Section 4.4 p 8059 line 7. “Other field campaigns”. Please give some of them.

These other field campaigns are discussed more later in the section. There is now a comment stating that there is further discussion below.

17. p. 8060, lines 5-6: “Meso-NH model does not include gas or aqueous chemistry”. I understand from section 3.5 that only the soluble species do not react. Are all the chemical species passive tracers in the model? If yes it should be written more explicitly in section 3.5.

Section 3.5 has been rewritten to clarify that gas and aqueous chemical reactivity is not included.

18. p. 8061, line 2: For HNO_3 , all the models except the RAMS model has anvil mixing ratios that are depleted...” Replace “has” by “have” It is an unexpected behaviour for a model which includes scavenging of soluble species. Is there a reason for this?

In the RAMS model the soluble species is scavenged via the Schwartz (1986) kinetic transfer equations. These equations result in rapid scavenging for small drops and much slower scavenging for larger drops (rain) Thus in the RAMS model, HNO_3 only gets into the condensed phase via the cloud drops. Since the production of rain directly from the cloud drops is small and the HNO_3 is degassed when cloud drops freeze, more HNO_3 is in the anvil region than found by other models. A sentence has been added

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to explain the RAMS result.

19. Conclusion: p. 8062 lines 14-22: again I think that the statement written here is too simple. A more detailed comment needs to be written here accounting for the questions mentioned above in this review. A comment on the potential impact of the microphysical scheme is also welcome. A sentence about what to do to properly simulate the possible intrusion of stratospheric ozone or other species would also improve the conclusion.

As discussed in the response to the referee's comments (above), a comparison of the lightning-NO_x parameterization brought out where uncertainties still exist. These uncertainties, with the recommendation of future measurements, are summarized in the conclusions. In order to remain focused on tracer transport in deep convection, the impact of microphysical schemes and the stratospheric intrusion are de-emphasized and therefore are not included in the conclusions.

20. About the need of field campaigns including measurements of soluble species: I fully agree with this. Can other recommendations for future campaigns be made here? For lightning NO_x parameterization?

A comment has been added citing the need for measurements of the NO source location.

Technical comment:

In Figure 6 panel d) please, change the Y axis so that all the model outputs can fit within the frame.

p. 8062, line 1: the year of the reference Cohan et al. is missing.

These changes have been made.

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