

Interactive comment on “A cloud chemistry module for the 3-D cloud-resolving mesoscale model Meso-NH with application to idealized cases” by M. Leriche et al.

Anonymous Referee #2

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This paper describes and evaluates a cloud chemistry module recently incorporated in the regional-scale Meso-NH model. The paper emphasizes the model description, which is well done, and results from three test cases. The test cases are interesting and appropriate because they begin with a liquid-only cloud and progress to a much more complex supercell thunderstorm. The results of the model simulation call attention to the importance of using a good cloud physics scheme for analyzing cloud chemistry processes. Further, continued work must be done to reconcile the role of the ice phase on cloud chemistry because, as the paper shows, the retention of trace gases when cloud drops freeze plays an important role in redistributing the trace gases. I have several minor comments and questions regarding the work. Once these comments

C172

and questions are addressed, I recommend the paper should be published.

Major comments:

1. One thing that is missing from this study is a way for model results to be easily compared with aircraft observations. I suggest that the authors include vertical profiles of the trace gas species in the convectively perturbed and unperturbed regions of the model.
2. While the authors discussed in the text the contribution of different processes to the trace gas concentration, it would assist the reader to show these contributions graphically. For example, vertical profiles of the different processes (e.g. the rate of transfer from cloud water to graupel, and photochemical production) would be useful.

Specific comments:

1. Section 2: How are photolysis rate coefficients calculated? Are they modified for cloud scattering? Are aqueous-phase photolysis reactions modified for increased path length?
2. Page 969, line 3. It would be good to show the actual polynomial equation that is solved to determine the pH of the drops.
3. Page 969, line 27. What effect does omitting gas trapping in growing ice hydrometeors have on the results? Does the Long et al. (2010) study quantify the contribution of retention via riming versus gas trapping (e.g., is gas trapping <10% contribution)?
4. Page 970, line 8. While the authors do say later (Section 3.3) that using one ice-phase concentration gives results of the same magnitude as those found by Barth et al. (2007), it would be nice to see a quantitative value describing differences between

C173

the method used in this study and a method predicting the trace species concentration in each cloud hydrometeor reservoir. Differences should arise because the fallspeed of snow and graupel are different and are even more different if hail is represented. Because this study uses the lower fallspeeds of graupel, it may be that representing the trace gas in ice with one prognostic variable is sufficient. This may not be true if hail is represented.

5. Page 971 and Table 4. The authors present the retention coefficients used for this study. Referee 1 commented that because of the importance of the retention process on the model results and the poorly constrained values of the retention coefficients, finding the sensitivity of the model results on the chosen retention coefficients would be useful to know. The referee also suggests making a recommendation for future studies based on the sensitivity analysis. I agree with the other referee that the sensitivity to the retention coefficient be analyzed. However, I would like to point out that there is not a consensus on the retention coefficients. Laboratory studies have been conducted finding a range of retention coefficients from 0.1 to 1.0 for a single species. As Stuart and Jacobson (2004) point out, there are several factors, e.g. Henry's law coefficient and how quickly the drop freezes, that contribute to whether a species is retained or not. Thus, I would recommend developing a physically-based method for assigning the retention coefficient (one that depended on these factors). Further, I would suggest that observations in real clouds continue to be made to help identify the retention coefficient, keeping in mind that drops that freeze quickly (possibly in severe thunderstorms) may have a different retention coefficient than clouds with drops that freeze more slowly (possibly in weak cold fronts).

6. Page 974, first paragraph. HARP case. While different initial profiles likely won't change the results of the study, I have a few questions and comments about the choice of initial profiles. Is the top of the boundary layer 1 km? Showing a profile of potential temperature would show the BL height and assure that the initial concentration profiles are consistent. Second, why is the stratospheric profile (O_3 profile) less at 3 km than

C174

at 2 km altitude? Most of my experience shows O_3 profiles to be fairly constant from 0 to 10 km altitude and then increase above this level into the stratosphere. Is there an ozonesonde profile from Hawaii that shows a typical vertical profile? Hydrogen peroxide has a distinctive vertical profile as well, with peak values occurring near the top of the boundary layer and lower values at the surface because of dry deposition and lower values above where less water vapor is available for H_2O_2 production. Have the authors done sensitivity simulations to explore the impact of the assumed vertical profile on the results of uptake and cloud chemistry?

7. Page 974, line 26. Is there significant production of H_2O_2 from aqueous-phase chemistry?

8. Page 975, lines 2-3. Could the authors explain better why the lower solubility of SO_2 delays its scavenging by cloud water compared to H_2O_2 ?

9. Page 975. Because trade-wind cumulus clouds grow and dissipate, it would be interesting to see what the redistribution of trace gases looks like after the cloud evaporates. Does the model simulation include this stage of the simulation and does it suggest interesting results e.g. a remnant cloud layer where formic acid exists, and SO_2 , H_2O_2 , and HCHO are depleted?

10. Page 978-979. The authors show HCHO mixing ratios for the COPT case and discuss processes that are important for those mixing ratios. These processes include sedimentation and melting of snow and graupel, and photochemical production of HCHO. It would help to show the contribution of these processes to HCHO using a figure. One way to do this is to produce vertical profiles of the horizontal sum of different processes affecting soluble tracers.

11. Pages 979-981. COPT case. Although it is not possible to compare these model results to aircraft measurements, it would be useful to plot the results in a way that could be interpreted for past and future aircraft studies. Often aircraft fly in the inflow

C175

region of the storm, get vertical profiles to the side of the storm, and fly in the outflow region of the storm. What would such a profile look like from these model results? I suggest showing vertical profiles that include typical inflow and outflow mixing ratios. Alongside this convectively-perturbed vertical profile, a vertical profile in the model domain that has not been affected by the convection would allow the reader to easily see the difference between convective outflow and background UT mixing ratios.

12. Page 979. COPT case. Even though the model results cannot be evaluated directly with aircraft measurements, it would be useful to know how these results compare to measurements taken in tropical squall lines, e.g. compared to the Borbon et al. (2012) paper.

13. Page 980. COPT case. How do the formic acid results compare to previous studies, e.g. Barth et al. (2007)?

14. Page 983, lines 26-29. STERAO case. It is not obvious in Fig. 11 that HCHO in the convective outflow is less than that in the unperturbed UT. Vertical profiles would help clarify the differences. In addition, the text simply says HCHO. Is it total HCHO mixing ratio or gas-phase mixing ratio?

15. Page 984. Same comment as above, but for the STERAO case. The authors show HCHO mixing ratios for the STERAO case and discuss processes that are important for those mixing ratios. It would be useful to show quantitatively the contribution of the processes via vertical profiles.

16. Page 984. A result that Barth et al. (2001) found for their simulations where $RET = 0$ for all species is that soluble trace gases that degassed when freezing occurred subsequently went into the remaining cloud water at the top of the updraft. Do the authors find this process occurring in their simulations?

17. Page 985, lines 10-12. What characteristics of the two microphysics schemes

C176

cause these differences? Different ice phase categories are mentioned, but I think the authors want to say that the higher density and fallspeeds of hail compared to graupel cause more dissolved species to transfer into graupel than hail. However, do the two microphysics schemes use different thresholds for when to create graupel or hail via a riming process?

18. Because I am requesting additional plots (see major comments), I suggest removing Fig. 13 because it looks like Fig. 11. That is, H_2O_2 behaves similarly to HCHO.

Technical comments:

1. page 964, line 8. I think the authors mean to say “mean molecular speed” and not quadratic.

2. Page 966, equation 6 needs the “c” subscripted for r_c .

3. Page 967, line 24. I think the authors mean to say, “chemistry mechanism”.

4. Page 967, line 28. Please add explanation of what is meant by “by first ordering the gas-phase species”. Does that mean the gas-phase chemistry was calculated first? Or does it mean that cloud-free grid points are calculated first?

5. Page 969, line 20. I think “generally” or “traditionally” is more appropriate than “classically”.

6. Page 970, line 1. Change to “pristine ice, which are newly formed. . .”.

7. Page 970, line 3. Change “concerned” to “included”.

8. Page 972, lines 6-8 should be written more explicitly. There should be a sentence saying that because species with $RET=0$ are not in ice, there is no need for a prognostic

C177

ice species equation. And line 6 could be written as “As shown in Table 4, 14 soluble chemical species and 5 intermediate ions have prognostic equations for the ice phase”. That is, clarifying early in the sentence what 14 and 5 refer to.

9. Page 973, line 18-28. I assume that the authors are reporting the findings by Cohard and Pinty (2000) throughout the paragraph. If so, it would be better to attribute their work at the start of the paragraph. For example, “Cohard and Pinty (2000) found numerous differences between They found after 1200 s of model simulation . . .”

10. Page 975, line 14. Please explain how Figure 6 is produced. Are these mixing ratios for a particular point in the model domain, or an average value for the model domain?

11. Page 976. It would be good if lines 10-11 are moved to the top of the paragraph.

12. Page 979, line 3. Spell out ETH and ALK for easier reading.

13. Page 980, line 26. Is the maximum value of 280 pptv correct (Fig. 9 has maximum values of 2 pptv)?

14. Fig. 5, pH cross sections. I suggest using contour levels of 0.2.

15. Fig. 6, Kessler is misspelled in the figure legend.

16. Fig. 10, The line segment in Fig. 10a is not placed correctly (it is too far to the left).

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