

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 #1

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The authors present a new sophisticated module of the well-known Meso-NH model. The new module includes both aqueous and gas phase chemistry. The model is applied to three exemplary cases that highlight different features of the multiphase system, i.e. one warm cumulus cloud case, comparing different microphysics schemes, a mixed-phase squall line and a supercell case. While in the first case the role of aqueous scavenging and chemical processes is highlighted, results in the other two cases suggest that retention of soluble species in ice might form an important reservoir and thus suppresses further chemical conversion in gas and aqueous phases. Overall the paper is well written and the new module is well explained. I have only some minor comments below that should be addressed prior to publication in ACP.

Main comments

1) Most of the retention coefficients are estimates and are chosen based on the species' solubilities. Since you show that retention is an important effect that either acts as a reservoir or as a means to transport species, I wonder how sensitive your findings to these values are. In case of high sensitivity, one conclusion of your study should be that more measurements of retention coefficients are needed in order to better constrain the partitioning of species between all phases.

2) Related to my first comments, it seems that the grouping of species in Table 4 is somewhat arbitrary. In particular the solubility of the acids will depend strongly on pH, and thus, some of the acids (HNO₂, HNO₄) might be only as soluble as CH₃OOH etc. At the very least, I think, you should add some comment to the Table that the different solubility categories are based on the assumption of a certain pH in order to avoid usage of these values in future studies for very different scenarios.

3) The Henry's law constant for HCHO listed in Table 1 is the effective Henry's law constant KH^* and includes hydration K_{hydr} , i.e. it is $KH^* = KH(1+K_{hydr})$ (cf Sander et al., 2006). Thus, including the hydration equilibrium in addition to KH^* , will lead to an overestimate of the solubility of HCHO. Is it possible that this overestimate can explain the discrepancy to the conclusions regarding HCHO transport and sources for Case 2 (p.979, l. 19ff).

Minor comments

p. 960, l. 12: This sentence seems out of place here and should rather be moved up to l. 2 where you talk about aqueous phase SOA formation.

p. 962, l. 10 f: Does 'chemistry' in these lines always mean the same, i.e. chemical reactions? Or does 'aerosol chemistry' refer to 'aerosol chemical composition'?

p. 963, l. 9: How are low-volatility aerosol constituents treated, e.g. sulfate, nitrate, ..? Are they kept completely in the particle phase upon drop evaporation? What fraction

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of sulfur is in the form of the sulfur-containing ions that are lost?

p. 969, l. 20:L What is meant here by 'ice reactivity'? Chemical reactions in ice? Or on the surface layer of ice?

p. 974, l. 12 and 18: Refer here to Fig. 2a and 2b, respectively.

p. 974, l. 27ff: Does your discussion indeed only refer to sulfur dioxide or to total S(IV)?

p. 977, l. 14: Is here the reference to a figure missing? If you do not intend to refer to a figure here, 'Results will be shown in the following' might be better.

p. 978, l. 19: Does 'scavenging' here also imply 'loss by reaction'?

p. 979, l. 24: The sources for formic acid are not really clear [Khare et al., 1999; Paulot et al., 2011]. The fact that in other model studies small gas phase contributions were found might be an artifact of the chemical mechanism.

p. 980, l. 9: do you mean 'net production' here?

p. 984, l. 22: Is there always sufficient SO₂ available to act as a significant loss for H₂O₂?

p. 986, l. 8f: This sentence is not clear. Please, reword.

p. 988, l. 11: It is true that a limited number of organics might lead to an underestimate of OH(aq) sinks; however, you also do not include several OH(aq) sources, such as Fenton reaction and nitrate photolysis. Overall, this omission might lead to partially cancelling effects and an approximately correct OH(aq) concentration.

Figure 2: It might help to add to the figures (not only in caption) 'gas', 'cloud', and 'rain'.

Figure 3: Is this total S(IV) or SO₂? If the latter, I would think that total S(IV) might be more meaningful.

Figure 6b: Some of the lines are hard to distinguish. I cannot find the one for 'SO₂ rain, C2R2' Technical comments

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p. 960, l. 5: better use 'e.g.', ' instead of 'for instance'

p. 961, l. 25: use consistently 'mixed-phase' or 'mixed phase'

p. 966, l. 10: equilibria

p. 968, l. 3. Does the sentence continue with 'e.g.'?

p. 979, l. 5: Better: 'small contribution' (the same at other places throughout the manuscript)

p. 981, l. 11: low mixing ratio

p. 985, l. 13: remove commas around 'in Meso-NH'

p. 985, l. 23: half of those

p. 985, l. 28: role of the ice phase

Figure 6: 'Kessler' misspelled in Figures.

Khare, P., N. Kumar, K. M. Kumari, and S. S. Srivastava (1999), Atmospheric Formic and Acetic Acids: An Overview, *Rev. Geophys.*, 37(2), 227-248.

Paulot, F., D. Wunch, J. D. Crouse, G. C. Toon, D. B. Millet, P. F. DeCarlo, C. Vigouroux, N. M. Deutscher, G. González Abad, J. Notholt, T. Warneke, J. W. Hannigan, C. Warneke, J. A. de Gouw, E. J. Dunlea, M. De Mazière, D. W. T. Griffith, P. Bernath, J. L. Jimenez, and P. O. Wennberg (2011), Importance of secondary sources in the atmospheric budgets of formic and acetic acids, *Atmos. Chem. Phys.*, 11(5), 1989-2013, 10.5194/acp-11-1989-2011.

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