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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The role of the particle size distribution in assessing aerosol composition effects on simulated droplet activation" *by* D. S. Ward et al.

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Response to Anonymous Referee #1:

We would like to thank the referee for their thoughtful comments that have significantly improved this work. All comments were addressed with sincere effort and we hope at a satisfactory level. Our responses to the comments and changes to the manuscript are included below in the order that the comments were given.

1) For the creation of the lookup tables for the RAMS droplet activation scheme I would suggest to improve the resolution of κ between 0.01 and 0.1. As it was shown in Reutter et al. (2009) (see Fig. 8) the sensitivity of the cloud droplet number concentra-

tions is largest for values below κ = 0.1 in all atmospheric regimes. Therefore a linear interpolation between 0.01 and 0.1 is maybe too coarse.

Reponse: This is an excellent point, especially considering the low values of κ (<0.1) that were estimated at SPL during ISPA-II. The parcel model was run for the entire parameter space at values of κ = 0.03 and 0.06 to increase the resolution between κ = 0.01 and 0.1. These results were used to construct additional lookup tables and installed in RAMS. The changes are indicated in the text.

2) Also for the choice of the RAMS sensitivity simulations I have the same suggestion to improve the number of simulations for κ values between 0.01 and 0.1, because here I would expect the greatest sensitivities (as it can be seen e.g. in the values for the spillover ratios in Table 3.).

Response: The RAMS sensitivity simulations were run for values of κ = 0.001, 0.03, 0.06 to investigate the sensitivity better in this range. Values of the spillover ratio for these new simulations are given in Table 3 of the revised manuscript. The results demonstrate the sensitivity of the spillover at these values of κ and also the lack of sensitivity for the higher values of median radius.

3) Page 4206, line 18 ff: Additionally of using the difference in precipitation between A1 and A7 the difference of runs within a realistic range of κ (e.g. 0.05 to 0.2) would be helpful. At least for the A-cases also a significant difference in the precipitation fields should be visible according to the spillover ratios given in Table 3 without an extreme κ range.

Response: With the results of the new RAMS sensitivity simulations recommended in comment #2 we created additional images showing the storm total precipitation difference for the more realistic range in κ as recommended. We used a range of κ = 0.06 to 0.20 which approximates the κ estimated from the ISPA-II observations in 2007. These images show very little sensitivity to the chosen range in κ even in the "A" simulations. This is an indication that the greatest changes in precipitation for the given storm occur for κ outside of the realistic range (probably below κ = 0.06). Evidence for this is also given in the spillover ratios shown in Table 3 of the revised manuscript. Above κ = 0.03 there is very little change in the spillover ratio, even for the A cases.

Since the images do not show sensitivity to changes in κ, we decided not to include them in a new figure but instead reiterated the fact that the realistic range in κ did not alter the precipitation pattern much for this case. This point was highlighted by the addition of the following text within the discussion of the spillover ratios:

P4207, L15: "In all simulation groups, the spillover ratio was apparently most sensitive to variations in the lowest values of κ, below the range of κ values estimated from the ISPA-II data."

4) As the authors report, McFiggans et al. (2006) state that the number of particles and the gradient of the size distribution determine CCN activity. Later the authors say that they kept the geometric standard deviation σg constant, because Antilla and Kerminen (2007) showed that moderate variations in the prescribed values of σg played only a minor role. Nevertheless, Antilla and Kerminen (2007) also showed that the shape of the particle mode could be as important as the mean size of the mode at low updraft velocities. Therefore I would be interested to see the sensitivity of the results on the geometric standard deviation, at least for the updraft-limited regime.

Response: The parcel model simulations were repeated for geometric standard deviations of 1.5 and 2.1 (original simulations used 1.8). The results are shown in Figure 2 of the revised manuscript. Discussion of the results was added at P4197, L27:

"The parcel model simulations were repeated for σ=1.5 and σ=2.1 to investigate the impact of the size distribution shape on these results. Antilla and Kerminen (2007) concluded that, for low updraft velocities, the size distribution shape can be just as important as the geometric mean radius for determining sensitivity to compositional changes. Here, values of S(κ) for the different distribution shapes are given by the dashed and dotted lines in Fig. 2. The narrowest size distribution (σ=1.5) shows the

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greatest values of sensitivity but trends the same as the other curves, as given by the dashed and dotted lines in Fig. 2. The values of S(κ) plotted in Fig. 2 also suggest that shape parameter variations are more important for small rg and no more or less important in the different regimes."

And at P4199, L22:

"The computed Pearson linear correlation coefficients, r = 0.80, 0.82, 0.85 for sigma = 1.5, 1.8, 2.1 respectively, suggest that the predictive ability of N(rc) is strong for the entire range of varied initial conditions, including median radius."

Note that we removed the original reference to Antilla and Kerminen (2007) and re-stated the reference in the new text to be more consistent with their conclusions as pointed out in this comment.

5) Fig.2: The authors should address the non-monotonic behaviour of the curve in Fig.2 c). What is the reason for local maximum of S (κ)? Reutter et al. (2009) reported problems with the model resolution for low supersaturations, which is the case for the updraft limited regime in Fig. 2c. Is it the same in this case?

Response: We had looked at the unexpected behavior of the updraft-limited curve as containing a local minimum instead of a local maximum but this comment helped us look at the problem in a different way. The following text was added to address this comment:

P4197, L25 "The non-monotonic path of the sensitivity curve in Fig. 2c could be the result of non-trivial variations in the difference between Smax reached for the lower κ limit and for the upper κ limit. Solution drops formed by more hygroscopic or larger particles remain in equilibrium with their environment at a lower saturation ratio, leading to the variations (Saleeby and Cotton, 2004). These variations in Smax increase and, therefore, become more important as rg increases. This effect, in combination with already enhanced sensitivity at larger values of rg in the updraft-limited regime

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(Fig. 2c), could have caused the apparent maximum in S(κ) near rg = 0.1 micron that occurred for all values of initial size distribution shape parameters. Of course, unstable behavior in the parcel model, which has been reported in such models at very low supersaturations (e.g. Reutter et al., 2009), could also be a factor."

6) Page 4193-4195: Model setup: It would be nice to have the technical details here at this place rather than a reference. Especially the time steps as well as the number of bins are interesting for an easier comparison with other studies. Also it would be nice to find the range of the initial values of T, w, Ncn and rg rather than in the reference of Saleeby and Cotton (2004). For example, was w really varied between 0.01 and 100 m s$-$1 ? Is this a realistic range for this studies?

Response: The timesteps, both external and internal, and number of bins used in the parcel model have been defined in the text. In addition, a new table was included (Table 1 in the revised manuscript) that includes all values of the 5 independent parameters that were used to initialize the parcel model.

It was also noted in this section that such large parameter ranges (such as the range in w pointed out in this comment) were needed because these results are used to construct the droplet activation lookup tables in RAMS. The parameter ranges need to be large enough to include all conditions that could be encountered in RAMS during the course of a simulation of various cloud regimes from supercell thunderstorms to marine stratocumulus. The large ranges used also add validity to the point made in section 2.3 by demonstrating that the relationship between S(κ) and rg applies for most observable conditions.

All the minor changes suggested were made. Thank you for your assistance with this manuscript!

Sincerely, Dan Ward and Coauthors

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 4189, 2010.