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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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 #2:

Thank you for your comments, especially those regarding ice-phase processes in this manuscript, which were not a focus of this study and yet are important in mixed-phase clouds. We have responded to all your comments here and hope that our edits and additions addressed them in a satisfactory way.

1) Section 2.1: Although appropriate references are given in the text some more information on the parcel model could be provided. Typically, parcel models do not consider sedimentation of hydrometeors. The water supersaturation reached in the model also

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depends on ice-phase processes such as the Bergeron-Findeisen process. The initiation of ice in a cloud in turns depends on heterogeneous ice nucleation. How are these processes treated in the model and how could they potentially affect the results? Some more discussion could be added here. It would also be helpful to summarize the parameter range used in the parcel model simulations in a table.

Response: This is correct, hydrometeor sedimentation is not simulated in this parcel model. The incarnation used in this study also does not consider ice-phase particles. Treatment of ice-phase and liquid-phase interactions in models is starting to become more common. In fact, the parcel model used here was upgraded recently to treat heterogeneous ice nucleation by Eidhammer et al. (2009). But we required liquid-phase only processes for the RAMS lookup tables and for comparison to the Reutter et al. (2009) results and the results of previous parcel model studies.

The following text was added to this section to make the absence of representation of these processes clear to the reader; P4194, L7:

"This simple air parcel representation captures the initial stages of liquid-phase cloud formation. The production of hydrometeors other than cloud droplets is not considered, nor are sedimentation processes that would remove water mass from the parcel."

Discussion of the possible impacts of neglecting these interactions is included in section 3.3 in also addressing comment #3. We added in the details of the bin number and timestep used in the parcel model simulations and also summarized the parameter ranges in Table 1 of the revised manuscript. This suggestion was also echoed by another referee.

2) Section 2.3: The authors cite McFiggans et al. (2006) and state that the CCN activity is determined by the number of particles and the gradient of the size distribution. Later the authors argue that variations in the geometric standard deviation play a minor role and refer to Antilla and Kerminen (2007). In light of the McFiggans et al. (2006) statement this argument seems counter intuitive. Some clarification is needed here.

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Throughout the study the geometric standard deviation is kept constant at σ=1.8. Why have the authors chosen σ=1.8? I suggest to either add a reference or back up this value with the observations. It would be interesting to see an additional sensitivity by varying the width of the mode within a reasonable range.

Response: These are all great comments. We removed the original reference to Antilla and Kerminen (2007) and restated the reference in new text (see below) to be more consistent with their conclusions as pointed out in this comment and that of another referee. The value for sigma of 1.8 was chosen to maintain consistency with the previous lookup tables used in RAMS and spelled out in Saleeby and Cotton (2004). This value can be considered to generally fall within the observed range, or really the range of values that result when lognormal distributions are fit to ambient aerosols. It falls within the range used by Antilla and Kerminen (2007), which was constrained by observations (although it falls a little on the high side of the range). These points are noted in the revised text. For the record, the average sigma of the lognormal curves fit to the many aerosol size distributions sampled at SPL during ISPA-II was 2.08, although this was likely skewed by the occasional presence of a small second mode at larger sizes.

We agree that it was an interesting question to vary sigma and investigate the sensitivity. 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 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

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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."

3) Section 3.3: One wonders why the authors have chosen an orographic mixed- phase case. From the presentation in section 2.2 one could have guessed a priori that the sensitivity would come out small because an aerosol-limited case has been investigated. With respect to the conclusions drawn from the model simulations I would argue that it is perhaps more interesting to look at an updraft- limited case (e.g., by modifying the initial conditions) and see if the conclusions still hold. Furthermore, I would argue that for precipitation from a mixed-phase cloud the composition effect is more important for the ice-phase (e.g., for heteroge- neous ice nucleation) than for the CCN activity (see for example Muhlbauer and Lohmann 2009). A short discussion in this direction would be helpful for clarifica- tion.

Response: A mixed-phase orographic cloud was chosen because this cloud regime has in the past been shown to be particularly susceptible to variations in CCN number concentration (Levin and Cotton, 2009 – Aerosol Pollution Impact on Precipitation). Surely the work by Muhlbauer and Lohmann (2009) and Muhlbauer et al. (2010 – ACPD) has shown that these clouds are complex and the susceptibility, even the sign of the precipitation perturbation, is still largely unpredictable. With that said, the specific case and model setup was chosen to mimic Saleeby and Cotton (2009) because they did show a strong spillover effect. Thus, the Feb. 2007 case really demonstrated the main idea laid out in the section 2, that changing rg can lead to changes in the sensitivity of a cloud to aerosol composition variations.

However, the point made in this comment is well taken. It is not necessarily appropri-

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ate to extend the conclusions drawn from this one case to similar clouds even in the same location because of possible differences in the phase distribution in the cloud and characteristics of the ice crystals. Along these lines, the generalizations made in this section were toned down to perhaps be more appropriate to the scope of this study. The following text was added P4207, L15:

“While ice-phase microphysics are treated in the RAMS scheme, the role of variations in ice crystals was not considered in this study. Recently it has been shown that these variations can influence the precipitation distribution in modeling studies of mixed-phase clouds. Muhlbauer and Lohmann (2009) introduced IN into idealized simulations of mixed-phase orographic clouds and found that increased number concentrations of IN can lead to an increase in total storm precipitation. When internally mixed aerosol are considered, particles are more likely to act as CCN than IN and the reduced riming described in the current study is reproducible. Still, the interactions between ice crystals and cloud droplets in these mixed-phase clouds are complex and still under investigation. These interactions may lead to variations in storm precipitation that are not predicted by the liquid-phase results put forth in this study.”

To address the first point (a little out of order), it is an interesting question to alter the initialization in RAMS to an updraft-limited case and compare to the aerosol-limited. In some respects this question exceeds the scope of this study for which the aim was to show how the sensitivity could be different within Reutter et al. (2009) regime for different aerosol size distribution. However, testing of an updraft-limited case would certainly help make this point even better. Here is where the choice of an orographic cloud is unhelpful.

There are two ways to re-initialize as updraft-limited: decrease w or increase Ncn. The former is difficult to do since we used a real case nudged to an observational dataset, but also because of the low-liquid water amounts characteristic of northwestern CO winter storms, to reduce the updraft speed to an updraft-limited regime value would almost certainly preclude the formation of the cloud. Ncn can easily be increased,

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so we re-ran RAMS simulations A1, A6, and A10 (see revised manuscript) with Ncn = 10,000 cm-3. The results showed very little sensitivity in the spillover to variations in kappa despite the updraft-limited conditions. This was likely because even at the low supersaturations there were so many particles present that a very low activated fraction (< 10%) still led to high CDNC and therefore, small mean droplet diameters.

Thus, the lack of sensitivity could be a result of the fact that the riming process has, in theory, a very sharp threshold droplet size for collection. So, we were unable to find sensitivity in an updraft-limited version of this case, but the result is still interesting and worth including. The following text was added:

"It is important to note here that because of the theoretically sharp threshold in droplet size that determines riming rate, the spillover will be greatest when changes in Nccn lead to droplet size changes above this threshold size. Cases for which Nccn is large will appear insensitive to variations in Nccn even for large changes in activated fraction."

4. P. 4207, l. 4-11: The definitions and explanations of the spillover should appear earlier in the text. E.g., moving the paragraph to p. 4206 after l. 5 seems more appropriate to me.

Response: Agreed, the spillover ratio text was moved to be included with the other spillover discussion.

All the technical comments were addressed. Thank you again for comments that genuinely improved this manuscript.

Sincerely, Dan Ward and Coauthors

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