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Atmospheric Chemistry and Physics
Discussions
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Interactive comment on "A comparison of two chemistry and aerosol schemes on the regional scale and resulting impact on radiative properties and warm and cold aerosol-cloud interactions" *by* Franziska Glassmeier et al.

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We thank referee 1 for the constructive and detailed comments and address each of them and corresponding changes to the manuscript below. Additional editorial changes are documented in a separately provided manuscript version with marked-up differences.

General comments:

1. Using (f1-f2)/(f1+f2) to see the percentage changes can be confusing and misleading, especially when f1 and f2 are quite different.

   In the submitted version of this manuscript we had defined the percentage change as $\Delta_{old} = (f_1 - f_2)/(f_1 + f_2)$ *because this expression has a nicer behavior for large relative differences than the version favored by the reviewer,* $\Delta_{new} := 2\Delta_{old}$. *For* $f_1 >> f_2 \Rightarrow \Delta_{old} = 100\%$ *such that* $-100\% < \Delta_{old} < 100\%$ *whereas* $-200\% < \Delta_{new} < 200\%$ *and* $\Delta_{new} = 100\% \Leftrightarrow f_1 = 3f_2$. *Despite this we agree with the reviewer that for smaller percentage differences* Δ_{new} *may be more intuitive and since both referees prefer* Δ_{new} *we have revised the manuscript accordingly.*

2. Suggest show both fields and then show the percentage differences.

   During our analysis we did not find distinct differences in the spatial patterns of the compared fields. Therefore additional panels showing the second field do not add additional insight as is illustrated in Fig. 1 below, which shows Fig. 3 from the manuscript with the MADE field included. As this can also be inferred by the absence of spatial structure seen in the difference plots, we decided not to include additional panels for the sake of clarity as the manuscript already contains a lot of figures.

3. I also suggest the authors add an acronym list at the end of the paper.

   A list of acronyms is added.

4. I also suggest the authors show the precipitation field from the simulation.

   As simulations with aerosol-cloud interactions are not meteorologically nudged simulations we deliberately abstained from showing precipitation fields. In order to attribute changes in precipitation to changes in aerosol treatment one would have to perform an ensemble of simulations in order to first capture the natural variability of the system and then to test if significant differences are found in the

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precipitation field exceeding the natural variability. Although this analysis would certainly be interesting it is beyond the scope of this study. The authors felt that showing difference plots in precipitation of the MADE and M7 simulations in comparison could be misleading. Such a plot may suggest to the reader that these changes could be attributed to the two different microphysics scheme, which is not necessarily the case.

Specific comments:

- Page 3, show full name of MADE:

  Changed accordingly.

- Page 4, Fig 1, caption. Show the meaning of the 2-letter abbreviations in the caption and add an acronym appendix.

  Changed accordingly.

- Page 4, line 13. If there is not nucleation scavenge, does impact scavenge take the hygroscopicity of aerosols into consideration? In another way, can increased SO4 coating affect the wet deposition of non-sulfate aerosols?

  page 5, line 13 has been changed for clarification:

  “The parameterization is applied to the wet aerosol radius such that the hygroscopicity of an aerosol particle may affect is scavenging efficiency by impaction.”

- page 5, line 14, how many insoluble modes in M7? seems 3 in Fig. 1

  Yes, there are only 3 insoluble modes. Changed accordingly.

- Page 7, How a PDF of updraft velocity is used in the activation scheme? Please add a few sentences to explain it.

page 7, line 17 has been extended for clarification:

“To take into account the sub-gridscale updraft variability, the number concentration of activated aerosol particles is determined by numerically averaging over a Gaussian probability density function (PDF) of updraft velocities about the grid mean value rather than using the number concentration of particles that are activated for the grid-mean updraft. The standard deviation of the PDF depends on the turbulent kinetic energy.”

- Page 8, line 20. Add “excess” in front of water vapor.

  The expression “excess water vapor” might be interpreted such that a saturation adjustment technique for the ice phase would be applied. This is not the case. The line has therefore been clarified in the following way:

  “[Ice-nucleation in ice-phase clouds follows the previous approach of MADE and] converts water vapor into ice.”

- Page 11, Line 7/8, why use “ca.” instead of “$\sim$” which is more straightforward?

  Changed accordingly.

- Page 11, last line, is the impaction scavenge scheme in the models able to describe such effect? If so, please add a few line describing the impaction scavenge scheme in the previous section.

  page 5, line 13 has been changed for clarification:

  “The description of impaction scavenging is based on an aerosol- and hydrometeor-size dependent collection efficiency. It considers inertial impaction and impaction from Brownian diffusion and interception but not phoretic effects.”

- Page 12, Line6, please clarify how you calculate the size distribution of internally mixed mode (like as,cs in M7) and how they are used to calculate the distributions of each species showed in Fig.5.

Page 16, fig 5. What is the “Total distribution” here, which curve? How are the distributions of each species calculated is not clear from the caption.

The caption of Fig. 5 has been clarified in the following way:

“Species include sea salt (SS), dust (DU), sulfate (SO_4), soot (BC) and primary organic carbon (POA). Individual lognormal modes are determined from the vertical sum and horizontal average of the the corresponding dry masses and numbers. The full, multimodal distribution emerges as the sum of individual log-normal modes. For mixed modes, lognormal modes of individual species are obtained by weighting the mixed-modal distribution by the fraction that the respective species contributes to the total mass in the mixed mode.”

- Page 12, line 16. From table 4, sea salt in M7 is 12% more (or ∼24% actually). Is this the mainly reason rather than being smaller?

  Yes, the text has been adapted accordingly:

  “[The M7 coarse-mode coating could be more effective because] sea salt is more abundant and particles are smaller such that a larger surface for condensation is available.”

- Page 13, Figure 3. Why use (f1-f2)/(f1+f2) instead of ((f1-f2)/(f1+f2)*0.5) to show the percentage changes. By using (f1-f2)/(f1+f2), you are essentially diminishing the percentage changes by a factor of 2 in all your discussions.

  See general comment (1).

- Page 14. Does BC in accumulation mode in M7 have more SO4 coating? If so, this may explain less BC in this mode via wet deposition.

  If the mixed M7 accumulation mode (as), which contains BC coated by sulfate, were to be scavenged more efficiently than the mixed MADE mode containing a soot core (jc), we would expect a decrease in M7 POA as well as BC, because

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these two species are internally mixed and thus always scavenged together. As we do not find this to be the case, we conclude that differences in wet deposition alone thus cannot offer an explanation for the differences in BC.

- Page 14, last line. does SO4 coating affect the impact scavenge?

  The current argument in the manuscript successfully explains differences in aerosol abundance between MADE and M7 based on differences in their dry radii and corresponding differences in deposition. The reviewer suggests that this argument might be incomplete because differences in the amount of sulfate coating will lead to differences in water uptake such that the wet radii may differ in a different fashion than the dry radii. We do not think that this the case for the following reasons: The M7 Aitken mode contains less SO4 than MADE, the M7 accumulation mode also contains less SO4 but more sea salt, the M7 coarse mode contains more sea salt and more SO4. For BC, the decreased SO4 coating strengthens our argument based on the dry aerosol radii instead of using the wet radii because it means even smaller particles due to decreased water uptake. For SS, we can assume that the SO4 coating is of minor importance as compared to the overall water uptake because sea salt is more hygroscopic than sulfate (kappa value of 1.12 as compared to 0.6 for sulfate). For POA, we can likewise assume that an increased water uptake due to the increased sea salt strengthens our argumentation made for the dry radius. Thus, although the amount of hygroscopic aerosol (sea salt and SO4) will control wet radii and deposition in an absolute terms, differences in dry radii seem to be a sufficient proxy for differences in wet radii, at least in our specific cases.

- Page 14, last line. Is there nucleation removal or other removal mechanism beside this?

  Nucleation scavenging is not considered in this study so that there are no other wet removal mechanisms.

- Page 15, line 2. Switch “4,3”. What is SIA?

  Changed accordingly and introduced meaning of SIA.

- Page 19, Line 5. Is SO4 coating from aqueous production of SO4 assigned to accumulation mode only or mainly? how is this determined?

  For clarification the following has been added to page 7, line 2:

  “SO4(aq) resulting from the aqueous-phase reaction is in most cases assigned to the mixed accumulation mode (mode as in Figure 1 and Table 1) and in fewer cases to the mixed coarse mode (mode cs). This is implemented by a number-based partitioning that favors the more numerous accumulation mode.”

  And the following to page 19, line 5:

  “[the aqueous-phase chemistry deposits sulfate mainly into the accumulation and to a lesser extent into the coarse mode] (the partitioning between these two modes is based on number and thus favors the more numerous accumulation mode)”

- Page 19, last 4 lines. What is the purpose of this compensation of sulfate burden in MADE ? Is it simply because M7 has more sulfate and MADE has more species? Need to justify why you compare it this way.

  For clarification, page 19, lines 7-10 have been rephrased as follows:

  “As discussed, M7 aqueous chemistry produces much higher sulfate concentrations, while MADE features nitrate and ammonium as addition aerosol species. Similar to different secondary organic aerosol (SOA) species, which are often lumped together, we combine sulfate, nitrate and ammonium into a secondary inorganic aerosol (SIA) class, to obtain a quantity that can be compared between MADE and M7. Note that for M7, SIA is identical to sulfate aerosol. From this perspective, the higher contribution of M7 sulfate to the total aerosol burden is compensated for by MADE nitrate and ammonium”

- Page 28, line 5. Suggest change CN=CCN, INP and N=. . .. Make it clearer.

  The text has been clarified in the following way:

  "The mediating effect of cloud microphysics on relative changes $\Delta N/N$ in a hydrometeor number concentration N that result from a relative change ΔCN/CN in the concentration of a cloud nuclei can be quantified [...] This universal equation quantifies liquid-phase (ice-phase) aerosol-cloud interactions when applying it to the droplet (ice crystal) number concentration N_{droplet} (N_{crystal}) and CCN (INP) by substituting $N = N_{\mathrm{droplet}}$ ($N = N_{\mathrm{crystal}}$) and CN=CCN (CN=INP)."

- Page 28, line 5. Also how is the CCN calculated? Use certain S or use the w* mentioned on page 10?

  page 10, line 21 has been amended for clarity:

  "In simulations coupled, the same updraft parameterization as used in simulations passive (i.e. no PDF) is applied for the online as well as offline calculation of CCN."

- Page 28, 26, "riming-splintering" has very narrow temperature range to apply. Is it possible to show the ice formation from this mechanism to the INP?

  Ice multiplication does indeed not seem to be the dominant mechanism to explain the high ice crystal concentrations as Fig. 2 below does not show a trend with temperature. To reflect this, the text has been adapted as follows:

  "Ice crystal number concentrations are always higher than the number concentration of INP in mixed-phase clouds (Figure 14, b). This can be attributed to ice crystal sources other than the heterogeneous freezing of cloud droplets. In our model, these are the freezing of rain drops, ice multiplication by rime-splintering and the sedimentation of ice crystals from aloft. These INP-independent ice crystal source processes [can also explain that crystal numbers are only weakly dependent on INP]"

- Page 29, last line. Please explain why larger aggregation number actually makes it closer to the 1:1 line

  We cannot identify a trend that larger aggregation numbers are closer to the dashed 1:1 line. They instead seem to increase with ice crystal concentration, i.e. along the y-axis of the plot.

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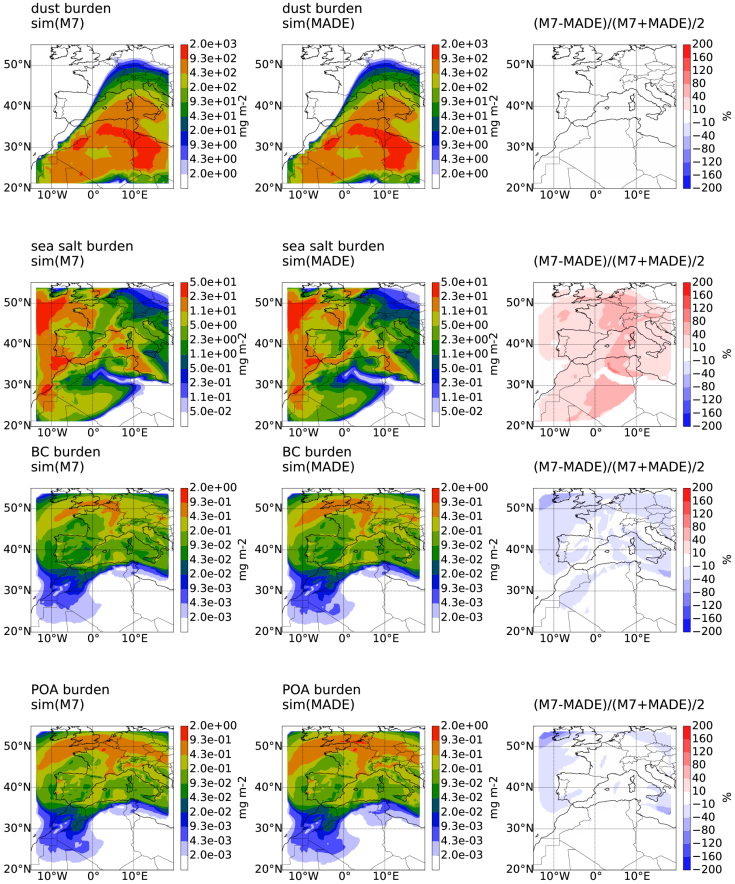

Fig. 1. Supplementary plot for Fig. 3: M7, MADE and percentage difference fields.

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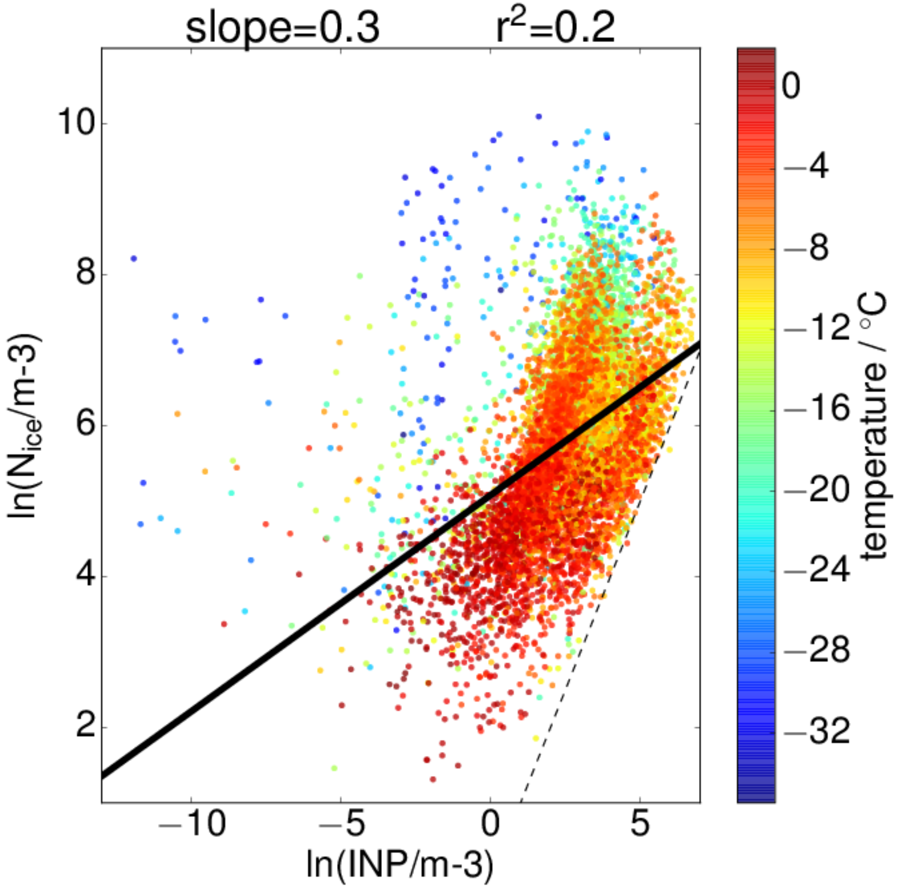

Fig. 2. Supplementary plot for Fig. 14: Role of temperature.