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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 2 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.

Major comment:

The author compared MADE designed for air quality applications and M7 for climate

projections in this study and concluded the importance of different processes or parameterization. However, one of the interesting information is not clearly presented and should be discussed. That is which processes are most important for air quality models and which processes are important for climate models, and possible improvement of future models based on your results.

We agree with the reviewer that this distinction is necessary for the clarity and direction of the manuscript. This discussion is now included in the revised manuscript on page 33, line 1ff as follows:

"We conclude that the new model version COSMO-ART-M7 simulates satisfying aerosol burdens in comparison to the established and observationally validated modeling framework COSMO-ART (Knote et al., 2011). Differences in burdens can be attributed to the choice of uncertain parameters, in particular modal standard deviation, and different structural assumptions in the form of missing species like SOA, nitrate and ammonium, and the choice of modes in terms of solubility and mixing state. This study provides the opportunity to discuss these choices in terms of the air-quality and climate objectives they are designed for. For climate applications, a computationally efficient aerosol scheme, such as M7, is needed that permits an as realistic as possible computation of radiative effects and aerosol-cloud interactions. As discussed earlier, simplified chemistry seems a viable option to save computational cost. In terms of aerosol-cloud interactions, the M7 approach to distinguish soluble from insoluble aerosol but to only consider one mixing state might be biased towards warm clouds. Ice nucleation not only depends on the mixing state of dust but also on an accurate representation of the dust surface. The latter is lost for internally mixed dust and soot. This raises the question if the representation of dust surfaces in M7 should be improved by following MADE in excluding dust from the mixed-modes and adding a separate, coated dust mode. To keep the original number of modes and the corresponding computational costs the same, the uncoated accumulation and coarse dust modes could be replaced by a coated and an uncoated dust mode of intermediate

size. When applying MADE for air-quality applications, the chemical speciation as well as the abundance of individual aerosol species, precursor gases and pollutant gases such as tropospheric ozone are of great interest. For this purpose, an as simplified treatment of chemistry as is used in M7 is no longer justified and a far more complex treatment is needed. However, this study confirms that an aqueous-phase chemistry that is efficient enough for the standard setup (a detailed aqueous-phase chemistry and wet-scavenging scheme for COSMO-ART has been developed by Knote and Brunner (2013)), may be a relevant objective for future model development."

Minor comments:

- Page 5: I suggest the authors to add a table to compare the detail of aerosol processes between these two aerosol modules.

  A new table (Tab. 2) has been added.

- Page 7 Line 3: Change 2.2.1 to 2.3, because Aerosol-radiation interactions are not relevant to 2.2 Sulfur chemistry.

  Changed accordingly.

- Page 8 Line 23: Why the author chose a Saharan dust outbreak reaching Europe in May 2008?

  Desert dust is one of the most effective natural INP currently known. Following Bangert et al (2012), we thus choose a dust outbreak to obtain a significant amount of ice-nucleation active aerosol so that we can analyze the difference between the two aerosol schemes not only in liquid clouds, but also in ice- and mixed-phase clouds. The following text is added to the manuscript for clarification:

  "Following Bangert et al. (2012), we choose a dust event to ensure sufficient INP concentrations inside our simulation domain in order to compare the implications

of aerosol schemes not only on liquid-phase processes, but also on ice nucleation rates in mixed-phase and ice clouds."

- Table: MADE passive simulation aqueous-phase chemistry and climatological oxidant fields. It may confuse readers if it shows 'y' as the same as M7. The authors should clarify it in table caption.

The caption has been clarified as follows:

"[A 'y' shows that a model feature is active,] if applicable (aqueous-phase chemistry and climatological oxidant fields are only active for M7 simulations and giant modes only apply to MADE simulations)."

- The authors used (f1 − f2) / (f1 + f2) to quantify relative differences in this study. I think the it should be (f1 − f2) / [(f1 + f2)/2] instead.

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.

- Page 19 Line 10: It may not be valuable to compare secondary inorganic aerosol between MADE and M7. Although M7 only simulate sulfate, the particle is actually (NH4)2SO4, NH4HSO4 in the atmosphere. Many climate model consider sulfate mass as NH4HSO4 instead of SO4. The author did not describe how is sulfate mass treated in M7.

The sulfate variable in M7 is interpreted as H2SO4 and not as NH4HSO4. This has been clarified on p5, line 18, as follows:

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"To be consistent with its simplified chemistry scheme, M7 sulfate is interpreted as sulfuric acid. M7 does not account for [nitrogen species and secondary organic aerosols.]"

- Page 28 Line 13: Because the authors are comparing two aerosol modules, it is better to show the figures for both of the two modules. Do they have the same information with the combined data?

The separate datasets have largely the same information (see Fig. 1 below). However, we do find some regime-dependence, which is most pronounced in the cirrus regime. Here, MADE and M7 feature very different levels of INP. The fit slopes obtained from both dataset together and for the individual dataset vary in the first decimal place. We assume that this variability gives an indication on the systematic error/accuracy associated to the fit slopes. Contrary to this accuracy estimation, the manuscript indicates fit slopes with two decimal places. We have corrected this by rounding the previous values of the fit slopes to one decimal digit in Fig. 14 and Tab. 8. The fit slopes reported in the manuscript are still obtained from the combined data set and not as an average of the two individual datasets. The results of both ways of averaging are practically identical (for the warm phase, combined fitting results in a slope of 0.8, whereas the average of the individual plots gives 0.85 and would thus round to 0.9) but combined fitting seems more accurate and keeps the manuscript shorter.

- Page 28 Line 16: Change smaller to lower.

Changed accordingly.

- Page 31 Line 13: Please clarify these conclusions are probably region-dependent. As I know, the aqueous oxidation of sulfate is sensitive over Europe, but some other regions are not.

p. 31, line 10 has been changed to:

"For this case, [a sensitivity study with identical emissions and identical parameterizations of dry and wet deposition for both schemes,] shows [the following sensitivities of simulated atmospheric aerosol burden, sorted in order of decreasing importance:]"

p. 31, line 19 has been changed to:

"It needs to be pointed out, however, that the importance of aqueous oxidation displays a strong regional dependence as it depends on cloud cover and droplet pH values."

• Page 31 Line 13: Also, please change chemical reactions to aqueous oxidation, because chemical reactions may mislead readers.

Changed to:
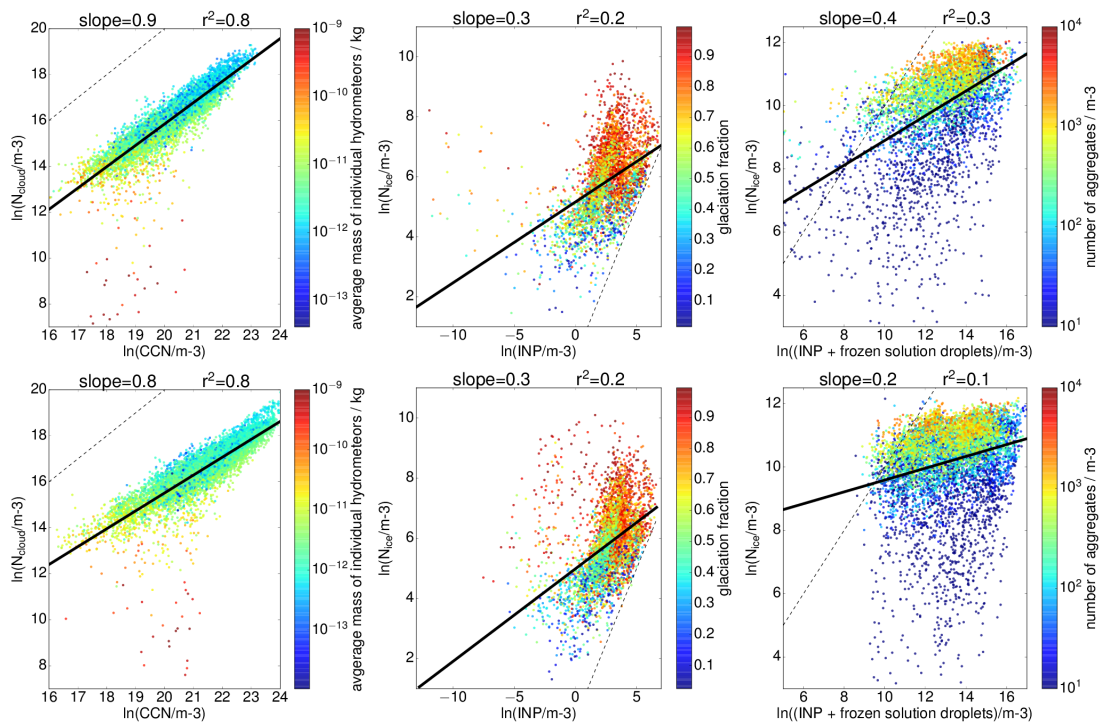
"consideration of sulfate production by aqueous oxidation"

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2016-1092, 2017.

Fig. 1. Supplementary plots for Figure 14: M7 (top) vs MADE (bottom) dataset.

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