

Reply to review by Anonymous Referee #1

We thank the referee for a thorough review of our paper and for pointing out critical aspects that required further clarification. We address (in upright font) the *comments* (indented and *in italics*) point by point below.

General comments

1. The authors report large deviations in number size distributions predicted by MADE3 and PartMC-MOSAIC in the diameter range critical for CCN activation and conclude that, “We will therefore have to be careful when interpreting CCN calculations in 3-D model applications.” This conclusion should be revised. If the deviations found here are likely to have a large impact on CCN concentrations, then further model development would be required for applications involving CCN. The authors should also indicate if the large deviations seen here are an artifact of the boxmodel approach (where transport, deposition, etc. are not included) and if these differences would likely be smaller in a 3D application.

Indeed, in the presented test case, there are large deviations in the number size distributions in the Aitken mode size range. However, since this test case is only one specific snapshot, and – as the referee correctly points out – several important processes were omitted here, only the 3-D application can reliably show if there are any systematic biases or not. The recent study by Mann et al., ACP, 2012, that compared a modal aerosol model to a sectional one within the same 3-D framework, indicates that some overestimation of CCN concentrations may be due to the modal approach in certain regions. Nevertheless, the choice of parameterization, in connection with environmental parameters such as the updraft velocity, has at least as large an influence on the results of activation calculations as the particle size (e.g. Ghan et al., JAMES, 2011; Simpson et al., GMDD, 2014).

We agree that the formulation of our conclusion as quoted above may be misleading. In the revised manuscript, that sentence is therefore replaced by the following two:

“Only the 3-D model application of MADE3, i.e. the inclusion of the processes omitted here and the consideration of many different combinations of environmental parameters and aerosol population states, will allow us to check against observations whether this deviation can be generalised and whether it could lead to a systematic bias in aerosol-cloud interactions. According to the results of a comparison of a modal aerosol submodel with a sectional one (Mann et al., 2012), CCN concentrations may be overestimated by the modal approach in certain regions.”

2. The Appendix compares size-composition distributions for MADE3 and PartMC-MOSAIC at the end of the simulation period, and the authors conclude that there is broad agree between the models. However, Figure A.1 appears to show some important differences. BC concentrations are elevated at small diameters in PartMC, and PartMC shows a decreasing trend in sulfate concentration with increasing diameter for fine particles. This behavior, which seems reasonable, is not captured by MADE3, but could potentially be important in many applications. The authors should do a better job of discussing differences in size-

composition distributions, even if it is difficult to perform a perfect 1-to-1 comparison between these models.

We acknowledge that a proper comparison of the different panels of Figure A.1 in terms of absolute mass concentrations required too many steps that were not shown: “sectionalizing” the MADE3 output, converting the fractional composition to absolute values, and observing the logarithmic scale of the mass axis. We therefore revised the figure (see supplement to this comment) by providing the “sectionalized” MADE3 output directly. In the new version, it is easier to see the broad agreement that we mention, especially in terms of the features that the referee points out. It is also important in this context to note that the decreasing trend in SO₄ with increasing diameter is in the fraction of SO₄ contained in the particles, and not in the absolute SO₄ concentration.

We revised the text of the appendix as follows, to reflect the changes in the figure and to address the referee’s comment:

“In Fig. A1 we show the initial and final states of the aerosol population as simulated by MADE3 and PartMC-MOSAIC. The figure illustrates the evolution of the aerosol composition under additional consideration of the size distribution. Composition is shown in a size-binned representation to facilitate the comparison of the individual panels. The top left panel contains the same data as Fig. 2 (note that the mass fractions of mineral dust are so small that they are not visible in Fig. A1).

The lower row plots in Fig. A1, representing the aerosol state after 24 h of simulated time, show the same general features: higher SO₄ fractions in the fine particles, predominant NO₃ partitioning to coarse rather than fine particles, and notable BC concentrations only in fine particles. Despite this agreement, one can also see that the modal approach leads to distribution of the components over wider size ranges. Since, due to their width, the modes contain particles of a broad range of sizes, this is inevitable in modal models.

The smallest particles do not take up water in PartMC-MOSAIC because they are assumed to be dry initially (Zaveri et al., 2005a) and the deliquescence relative humidity of (NH₄)₂SO₄ is higher than the environmental relative humidity specified in our experiment (0.771). Conversely, in EQSAM, these particles do take up water due to the presence of small quantities of other components that reduce the particles’ deliquescence relative humidity.”

3. In several places in the manuscript, the use of different thermodynamic modules in MADE3 and PartMC-MOSAIC complicates the interpretation of differences in model predictions. If the authors were to re-run the test case using the MOSAIC thermodynamic module within the MADE3 formulation, it would help separate differences in model predictions that are due to aerosol dynamics and thermodynamics.

We totally agree with the referee here and we would like to implement MOSAIC as a MESSy submodel in the future to enable its use with MADE3 (and other aerosol submodels). However, this is beyond the scope of the current work. We intend to determine how the inclusion of the interactive coarse mode changes the conclusions from previous studies. Since these prior simulations were all conducted using EQSAM, we refrain from changing the thermodynamics code at this point. We hope the referee will understand our decision, and added a note of caution w.r.t.

this difference to the model description of PartMC-MOSAIC (section 2.3) at the end of l. 10 on p. 711:

“Note that, besides the different approaches to aerosol microphysics, the use of different codes for the thermodynamic calculations (EQSAM in MADE3 vs. MOSAIC in PartMC-MOSAIC) can be a major driver of differences in simulation results.”

4. The authors considered only one boxmodel test case (marine conditions) because their initial 3D application will focus on shipping impacts. However, MADE3 will likely be applied under diverse conditions in the future. Including additional test cases (e.g., mixing of urban and marine air masses near the coast) could also be informative.

Actually, we did also simulate other test cases, but decided not to discuss them in the paper for two reasons. Firstly, the main goal of this work was to test the newly introduced interactions of coarse mode particles with the condensable gases and the fine particles. This requires a test case with a significant amount of coarse particles, preferably sea spray particles to also test the newly included HCl/Cl partitioning. Hence, the one presented here was an obvious choice. Secondly, the general conclusions from the other test cases were the same as the ones presented. For individual species, there were different deviations that could be explained by the different thermodynamic codes, mainly by the different approaches to compute activity coefficients. Hence, we did not consider the additional test cases to add any value to the article. Furthermore, as noted above, many more “test cases” will be implicitly included in the evaluation of MADE3 within the 3-D model. Nevertheless, we would like to point out that the deviations in the fine particle size distributions were not as large in the other test cases as in the one presented.

Specific comments

–Abstract: Please clarify that the model described here is MADE3v2.0b (if that is the correct version). There could be confusion because the article appears to describe the beta version of version 2 of the 3rd generation of the MADE model.

Both is actually correct, as we named the 3rd generation of the MADE submodel 'MADE3'. We added this piece of information to the first parenthesis in the abstract:

“We introduce MADE3 (Modal Aerosol Dynamics model for Europe, adapted for global applications, 3rd generation; version: MADE3v2.0b) [...]”

–Abstract, line 23: Please clarify that “total aerosol composition” is being referred to (size-composition distributions appear to differ significantly between the models).

They do not actually differ very strongly (see the response to general comment 2), so that we did not modify the text here.

–Section 2.1.2: Please clarify if H₂SO₄ can condense onto insoluble BC/Dust distributions

We added the following sentence after the motivation for the choice of alpha_H₂SO₄:

“The value is the same for all modes, i.e. condensation is treated in the same way, regardless of

whether particles contain insoluble material or not.”

–p. 701, line 21: In this manuscript, is “coarse particles” used to indicate the large diameter modes?

The point is well taken, we used the term “coarse” in several different contexts. We took care now to clarify what we mean throughout the manuscript, replacing “coarse particles” by “coarse mode particles” or “coarse particles (sizes $\geq 2 \mu\text{m}$) ” etc. where appropriate.

–p. 701, line 27: You should probably add the Sun and Wexler (1998, AE) reference for completeness

Sun and Wexler, in their 1998 AE paper, describe a method that takes advantage of special features of the aerosol along the south-western coast of the U.S. (acid neutrality). It is therefore not generalizable for application in a global model, so that we feel we should not cite it in the context of MADE3. We hope the referee will agree with our decision.

–p. 702: Please clarify why H₂SO₄ is being treated dynamically when the previous page indicates that time scales justify an equilibrium approach and equilibrium is assumed for other components. Is this related to the need to treat nucleation for H₂SO₄?

Yes, it is related to the need to treat nucleation, but that is not the only reason. As measurements suggest a negligible amount of gaseous H₂SO₄ to be present in the atmosphere, we assume that all H₂SO₄(g) that is produced within a time step is transferred to the aerosol phase. If we employed the equilibrium approach here, the amount that would have to condense might exceed the maximum amount that would be physically possible during one time step (due to the diffusion limitation, as discussed for the other condensable gases and large particles), even for fine particles. Furthermore, the equilibrium approach would not allow for a correct distribution of H₂SO₄ among the different modes. Therefore, we calculate the condensation flux explicitly and perform the nucleation calculation if condensation alone cannot remove all the H₂SO₄(g), as stated in the paper.

To clarify this, we added a few introductory sentences to the section on H₂SO₄ condensation (the following replaces the sentence leading to Eq. 7):

“Due to its very low equilibrium vapour pressure, we assume that all H₂SO₄ is transferred from the gas phase to the aerosol phase during each time step. Depending on the magnitude of the condensation flux, this transfer can occur via condensation alone, or via condensation and new particle formation (see below). To determine the amount of H₂SO₄ that can condense during one time step, we calculate the condensation flux explicitly. This is also necessary to obtain the proper distribution of the condensate among the differently sized particles. An equilibrium assumption does not yield this distribution. While potential errors in the size distribution of condensing material will be corrected by re-evaporation in case of the semi-volatile species, this is not possible for H₂SO₄, since our assumption that all H₂SO₄ is transferred to the aerosol phase means that it cannot re-evaporate in our model. The total condensation flux of H₂SO₄ is the sum of the rates of change of mass concentrations $c_{\text{H}_2\text{SO}_4,k}$ for all modes ($k = 1, \dots, 9$): ”

–p. 711, line 24: The phrase “coarse particle interactions” is used in various places in the

article. Please clarify what is meant in the context of marine conditions. For example, is the issue water competition during activation, effects of giant CCN on autoconversion, etc.? Also, the manuscript seems to imply that the replacement of Cl by NO₃ could be important for water uptake and activation. Is this so? I would think that soluble coarse particles would easily activate regardless of whether they contained NaNO₃ or NaCl.

The phrase “coarse particle interactions” refers to interactions of coarse mode particles with fine particles and condensable gases throughout the paper. We always refer to the same feature with this term, namely the main new feature of MADE3. To clarify this, we added “with fine particles and condensable gases” or similar phrases where appropriate.

Concerning the Cl/NO₃ issue, the referee probably refers to the sentence on p. 720, ll. 26ff. We agree that this could be taken to imply that the Cl replacement by NO₃ could lead to reduced water uptake and thus affect activation. What we intended to say in this sentence only concerned Cl, however, not NO₃. Therefore, we rephrased it to:

“The evaporation of some of the Cl to the gas phase (as HCl) may entail differences in aerosol water content if it is not fully replaced by NO₃. This, in turn, may affect [...]”

–p. 712: It might be helpful to include a table of initial conditions for the model simulations in case other groups would like to repeat the test with their models.

Following the referee's suggestion, we added a table with the initial species mass and number concentrations, aggregated by size ranges, to the setup section.

–p. 717, line 18: As the authors are probably aware, it is possible to simulate the evolution of particle mode standard deviations dynamically. Models such as CMAQ currently do this, although they include limits on the range in which the standard deviations can vary, and so it is unclear how numerically stable these calculations are.

Indeed, we are aware of such approaches that predict all three moments of the log-normal size distribution instead of fixing one. It would be interesting to include a model that predicts all three moments in a comparison like the one presented here. However, it is beyond the scope of the current work to test whether such a scheme would bring the MADE3 results closer to the PartMC-MOSAIC size distribution. For further justification of the fixed mode widths in MADE3, see Dr. Binkowski's review.

–p. 719, lines 9-10: Does this mean that coarse particles components were effectively in equilibrium with the gas phase in MADE3 in this test case?

It means that the model treated the coarse mode particles as if they were in equilibrium with the gas phase, and it means that they were not very far from equilibrium. However, as the flux limit that we apply is quite “generous”, we would not state that they “were effectively in equilibrium”.

References

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