J. C. Kaiser, J. Hendricks, M. Righi, N. Riemer, R. A. Zaveri, S. Metzger, and V. Aquila:
The MESSy aerosol submodel MADE3 (v2.0b): description and a box model test

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 paranthesis 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 >= 2 µm) ” 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_H2SO4,*k* for all modes (*k* = 1,...,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 equlibrium 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

Ghan, S. J., Abdul-Razzak, H., Nenes, A., Ming, Y., Xiaohong, L., Ovchinnikov, M., Shipway, B., Meskhidze, N., Xu, J., and Shi, X.: Droplet nucleation: physically-based parameterizations and comparative evaluation, J. Adv. Model. Earth Syst., 3, M10001, doi:10.1029/2011MS000074, 2011.

Mann, G. W., Carslaw, K. S., Ridley, D. A., Spracklen, D. V., Pringle, K. J., Merikanto, J., Korhonen, H., Schwarz, J. P., Lee, L. A., Manktelow, P. T., Woodhouse, M. T., Schmidt, A., Breider, T. J., Emmerson, K. M., Reddington, C. L., Chipperfield, M. P., and Pickering, S. J.: Intercomparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model, Atmos. Chem. Phys., 12, 4449–4476, doi:10.5194/acp-12-4449-2012, 2012.

Simpson, E., Connolly, P., and McFiggans, G.: An investigation into the performance of three cloud droplet activation parameterisations, Geosci. Model Dev. Discuss., 7, 1317–1336, doi:10.5194/gmdd-7-1317-2014, 2014.

Sun, Q. and Wexler, A. S.: Modeling urban and regional aerosols - Condensation and evaporation near acid neutrality, Atmos. Environ. 32, 3527–3531, doi:10.1016/S1352-2310(98)00059-4, 1998.

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Reply to review by Dr. Frank Binkowski

We thank Dr. Binkowski for his encouraging words and for his comments that helped to improve our article. They are answered (in upright font) point by point below (*comments* indented and *in italics*).

> *Discussion of coagulation is not quite transparent. For example Equations 1 and 2 describes the number coagulation process from Riemer (2009), but equations 13 and 14 are for number and mass and seem to have little to do with equations 1 and 2. Further the discussion of equation 15 is not completely clear because it depends upon the first Kroecker delta in which the subscripts are not quite consistent; e.g. one subscript is an integer, the second subscript is an array. The coagulation in this system is complicated and important. Thus, an appendix describing this in a bit more detail would help the reader grasp the way coagulation is actually calculated. Further, a diagram or table similar to Table 3. of Aquila et al. (2011) for the 9 by 9 system described here would be greatly appreciated.*

We agree that our formulation of the coagulation equations (13 and 14) is not easy to grasp at first glance. However, we could not find a more concise and at the same time complete formulation. Following Dr. Binkowski's suggestion, we added a coagulation target mode table to Section 2.1.2. In the revised manuscript, we swapped the examples and the rules for the target mode matrix elements (tau_lm), and added some further explanations to the examples in order to clarify the connection of the equations with the matrix:
“For instance, particles that result from intermodal coagulation of particles from modes l = 1 (‘ks’, or soluble Aitken mode in Table 2) and m = 4 (‘as’, or soluble accumulation mode) are assigned to mode 14 = 4 (‘as’). Hence, a_14^4 = 0, which means that this process does not add to the particle number in mode k = 4 (‘as’). It does, however, add mass from mode l = 1 (‘ks’) to mode k = 4 (‘as’). This is reflected in the parentheses with the Kronecker symbols in Eq. (14): the first parenthesis evaluates to one, the second parenthesis to zero. In case of intramodal coagulation, i.e. if l = k and m = k, the value of the coefficient in Eq. (13) is a_kk^k = -0.5. It is negative because one particle per such event is lost, but the factor is only -0.5 because of the double integration over the same mode. In this case, all the Kronecker symbols in Eq. (14) evaluate to one, so that all summands are zero, and no mass is added to mode k.”
This should also show more clearly that all Kronecker deltas actually have two integers as their subscripts. While tau is a matrix (array), tau_lm represents the matrix element with the coordinates l and m, which is the (integer) number of the target mode for the coagulation of particles from modes l and m.
We also considered adding an expanded example set of coagulation equations (i.e. for number and mass) to the appendix, with the sums and Kronecker deltas written out explicitly for one mode. However, as the resulting equations filled more than a whole page, we decided to leave them out and only added the following note on the number of non-zero terms (after the examples as given

above):
“In total, both Eq. (13) and Eq. (14) include 45 summands for each mode k, but many of them are zeros. For example, coagulation losses and gains in the soluble coarse mode (cs in Table 2) are described by 7 non-zero terms in the number equation (Eq. 13), and 8 non-zero terms in the mass equation (Eq. 14).”
Concerning the overall presentation of the “aerosol mathematics” we have to slightly disagree with the comment. Equations (1) and (2) fully describe the evolution of the aerosol population (as simulated here). Both number and mass enter this equation as n*(mu, t) is the number concentration “per particle composition” (note that the mu_a are the particle component masses). We therefore consider this a very general form of the aerosol dynamics equation, which – in our opinion – justifies its use in the introduction of the model description. As MADE3 relies on log-normal functions in diameter space, we have to relate both number and species mass concentrations to the diameter. This leads to the split-up of the coagulation terms in Eq. (1) in one for the number concentrations (Eq. 13) and one for the species mass concentrations (Eq. 14). The general form of the three equations, i.e. the integration over the product of the coagulation kernel and two number distributions, is the same.
We hope that this explanation makes our choice of how to present the aerosol dynamics equations plausible.

> *Page 709. The authors chose to compare MADE3 with MADE from the Lauer et al. (2005) paper rather than comparing with MADE-in (Aquila et al., 2011) even when they say the path to MADE3 was through MADE-in. This confuses the situation. MADE3 is closer to MADE-in than it is to MADE. Why not just compare with MADE-in?*

As stated in the introduction, this choice is due to the first target application of MADE3, namely the re-assessment of the shipping effect on the global aerosol. We intend to perform a study similar to Lauer et al., ACP, 2007; AST, 2009, and Righi et al., EST, 2011; ACP, 2013, which were all conducted using MADE. The effect of including the interactive coarse mode will be assessed by comparison with the latter studies. Hence, we also chose to compare MADE3 to MADE here, in order to have a reference. In our opinion, the relatively small differences that we found in the fine particle size range corroborate the validity of such a comparison.

> *Page 710 –lines 1 and 2 How do dust particles get into the Aitken mode?*

This point is well taken, and the answer is: they do not. We meant to say that, from a technical point of view, all species could now be present in each mode, to explain the number of species mass tracers in MADE3 vs. that in MADE: 81 (= 9 modes times 9 tracers) vs. 18 (does not equal two fine modes with eight tracers each, plus three tracers in the coarse mode). If we do not assign dust emissions to the Aitken mode (which indeed we do not), this tracer's value will just be zero at any time. We removed the confusing sentence, because it relates only to the technical implementation (2-D arrays for tracers in MADE3 vs. 1-D arrays in MADE).

> *Pages 710 and 711 describe PartMC-MOSAIC and alternative aerosol dynamics method that tracks individual particles rather than particle distributions. (Here is where Equation 1 is more appropriately displayed). The authors have chosen to use PartMC-MOSAIC as a*

> *reference method. It is important to note here that the difference between MOSAIC and EQSAM (EQuilibrium Simplified Aerosol Model) form a second confounder between MADE3 and PartMC-MOSAIC. Thus, comparisons between MADE3-EQSAM PartMC-MOSAIC should be made with great caution, as the authors have done.*

We followed Dr. Binkowski's advice to add a note of caution in the PartMC-MOSAIC description. For the wording, please refer to our reply to the anonymous reviewer’s comment (suggestion to use MOSAIC instead of EQSAM with MADE3).

> *The difference in Aitken mode behavior between MADE3-EQSAM and PartMC-MOSAIC is most likely due to the fact that these comparisons are done in a box-model. Transport processes tend to broaden the size distributions in an Eulerian framework. This broadening is often excessive as our experience with CMAQ shows. The limitations that we put in were to keep the geometric standard deviations from growing too large.*

We thank Dr. Binkowski for sharing his experience with variable geometric standard deviations. It would be interesting to see if PartMC-MOSAIC would also show broadening of the size distribution in a 3-D transport enabled model. However, as available computational power currently does not permit productive usage of PartMC-MOSAIC within a 3-D model, such experiments will have to be postponed to the future.

> *Ambient data do not seem to show this very narrow Aitken mode. Therefore, fixed geometric standard deviations is a much better idea 3D codes.*

The very narrow Aitken mode is certainly a feature specific to our setup (with transport and deposition neglected). In the real world, such narrow modes will likely be rare, although they could exist, for instance, in close proximity to emission sources. However, as global 3-D chemistry climate models do not resolve such small scales, we also feel that fixed mode widths are a valid simplification for such applications.

References

Lauer, A., Eyring, V., Hendricks, J., Jöckel, P., and Lohmann, U.: Global model simulations of the impact of ocean-going ships on aerosols, clouds, and the radiation budget, Atmos. Chem. Phys., 7, 5061–5079, doi:10.5194/acp-7-5061-2007, 2007.

Lauer, A., Eyring, V., Corbett, J. J., Wang, C., and Winebrake, J. J.: Assessment of near-future policy instruments for oceangoing shipping: impact on atmospheric aerosol burdens and the Earth’s radiation budget, Environ. Sci. Technol., 43, 5592–5598, doi:10.1021/es900922h, 2009.

Righi, M., Klinger, C., Eyring, V., Hendricks, J., Lauer, A., and Petzold, A.: Climate Impact of Biofuels in Shipping: Global Model Studies of the Aerosol Indirect Effect, Environ. Sci. Technol., 45, 3519–3525, doi:10.1021/es1036157, 2011.

Righi, M., Hendricks, J., and Sausen, R.: The global impact of the transport sectors on atmospheric aerosol: simulations for year 2000 emissions, Atmos. Chem. Phys., 13, 9939–9970, doi:10.5194/acp-13-9939-2013, 2013.

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List of relevant changes in the revised manuscript

Besides the changes mentioned in the replies to the reviews, we made the following changes to our revised manuscript (note that none of these modifications affect any of the results or conclusions presented in our article):

- PartMC-MOSAIC panels of Fig. A1: we detected a small error in the processing of the PartMC-MOSAIC output data that led to an incorrectly represented size distribution of SO4. This is fixed in the revised version.
- Description of how we determined the gaseous sulfuric and nitric acid production rates (Table 1): in the GMDD version we had accidentally included an outdated version of this description. We therefore replaced it in the revised manuscript, so that the description is now consistent with the values of the production rates as given in Table 1. Note that these values were not changed with respect to the GMDD version.
- We changed small parts of the text in some cases where we thought it would help the reader to understand our article more easily.
- We fixed a few typos.