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

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