**Geoscientific
Model Development
Discussions**

Interactive comment on "MADE-IN: a new aerosol microphysics submodel for global simulation of potential atmospheric ice nuclei" *by* V. Aquila et al.

V. Aquila et al.

valentina.aquila@nasa.gov

We thank the reviewer for the useful comments. We agree that our focus is not the simulation of ice nuclei (IN). The simulation of IN is indeed not possible, since it is still unclear which microphysical and chemical properties enable only a specific fraction of mineral dust or BC particles to act as efficient IN. We aim at simulating the properties (number, composition and mixing state) of insoluble particles which could possibly act as IN. We intend to use this information to assess possible effects of IN on cirrus clouds by coupling MADE-in to an ice nucleation parameterization (e. g. Kärcher et al., 2006) within the global model framework. To this end, the fraction of BC and dust particles which could actually act as ice forming nuclei as well as other relevant parameters, such as the respective humidity thresholds, have to be defined accordingly to drive the

ice nucleation parameterization. The information about the mixing state can be used to separate potential deposition nuclei from immersion nuclei and to assume different nucleation abilities for these different IN types. We agree that the name of the model might be misleading, and that this argumentation was not clear enough in the previous version of the paper. We have modified the manuscript accordingly. We have generally shifted the focus of the paper from ice nuclei aspects towards insoluble particle issues. The name MADE-in now stands for modal aerosol dynamics model including insoluble modes. Due to the change of focus of the paper, the following reviewer comments, which regard the possibility of using MADE-in to study IN, are less relevant.

1. **Reviewer comment (RC):** There is contradictory evidence on whether coatings suppress ice nucleation, have no effect or even enhance it. According to some studies, immersion freezing is the most likely ice nucleation mode in mixed-phase clouds. If activated to cloud droplets, however, the soluble material of the coatings would be so diluted that the effect on the freezing efficiency is probably small, unless the particle surface has been modified by chemical reactions.

   Authors reply (AR): We agree that there is contradictory evidence on the effect of the coating on ice nucleation, particularly in the mixed-phase cloud regime. In the cirrus regime, coatings may dampen the ice-forming ability of mineral dust (e. g. Möhler et al., 2008; Cziczo et al. 2009; Koehler et al. 2010). In the case of BC at cirrus temperatures, the information is also contradictory (DeMott et al., 1999; Möhler et al., 2005; Käercher et al., 2007). EMAC/MADE-in calculates the number and mass concentration of internally and externally mixed insoluble particles, but does not make any assumption on their freezing ability. This should be done within an ice microphysical scheme, once coupled to EMAC/MADE-in. Even though the effect of the coating on ice nucleation is not clear yet, a model like EMAC/MADE-in allows to perform sensitivity studies with various scenarios, assuming that the internal mixture facilitates or not the formation of ice crystals.

2. **RC:** To the extent of my knowledge, there is currently no ice nucleation parameterization available which would make explicit use of the information on the mixing state and coating thickness.

   AR: As stated in the general comment above, information about the chemical composition and mixing state of ice nuclei can be used to identify which freezing process is more likely to take place.

3. **RC:** Particles other than dust and BC have been identified as ice nuclei, including organic acids and humic-like substances, metallic particles and bioaerosols. These are not even mentioned.

   AR: We agree that BC and dust are not the only ice nuclei in the atmosphere, and other species as organic aerosol or metallic particles appear to be very effective ice nuclei. With regard to the cirrus cloud regime, the major research activities focused on mineral dust and BC IN and this is the reason why these particle types are in focus of MADE-in. However, the insoluble particle modes of MADE-in can be extended by other components in future studies.

4. **RC:** "Potential IN" is a term invented by modellers and is not well defined. What measurable quantity does this correspond to? Does this refer to all particles which can induce heterogeneous above a certain temperature? If so, above what temperature? You should note that in no laboratory of field experiment, more than a at maximum few percent of all BC particles were found to nucleate ice, even at low temperatures. (This is different for mineral dust.)

   AR: We have omitted the term potential IN in the current version of the manuscript.

5. **RC:** DeMott et al. (2010) showed a remarkable correlation of IN with concentrations of aerosol particles larger than 0.5 μm. This points to a first-order dependence on size rather than on mixing state.

<div align="center">C880</div>

   AR: Since EMAC/MADE-in also resolves the size distribution of insoluble particles, also this issue can be addressed.

6. **RC:** The simulated dust concentrations are not properly evaluated. This is crucial if this model is supposed to be used for simulating IN. The coarse mode receives much less attention in this manuscript than the Aitken and accumulation modes, although it might be the most important size class for potential ice nuclei. Coating of the coarse mode should be included in the model, as this can be a major sink for condensable gases.

   AR: We have included an additional evaluation of mineral dust concentrations at the Earth's surface by comparing with the station data discussed by Huneeus et al. (2010). We agree that coarse mode dust particles can be important ice nuclei, but their large size mostly keeps them confined to lower altitudes. This is now mentioned in Sect. 2.1 of the manuscript. Since we plan to apply EMAC/MADE-in to study cirrus clouds, we mainly focus on sub-micrometer aerosol.

7. **RC:** For the purpose of simulation of IN, I see a major problem in the fact that the "acc$_{\mathrm{ext}}$" and "acc$_{\mathrm{mix}}$" modes contain both BC and dust, such that the number concentration of dust particles can not be diagnosed. One eventually has to assign different ice nucleation abilities to BC and dust, but here they are not treated separately. Probably the "potential IN number concentrations" in Fig. 16 are dominated by BC. How would the dust number concentrations look like?

   AR: EMAC/MADE-in does not simulate separately the number concentration of BC and dust, but does simulate their mass. To drive ice nucleation parameterizations, the number contributions of BC and dust could be diagnosed from their mass contributions to each mode. This however will be the subject of future studies as described in our general remarks above.

Detailed comments.

<div align="center">C881</div>

1. **RC:** Introduction: As already pointed out by Daniel Cziczo, the introduction should contain a more accurate representation of the present state of knowledge on heterogeneous ice nucleation.

   AR: The introduction has been modified. The focus has been shifted from ice nucleation to the importance of insoluble particles and their mixing state in general, including some more details about ice nuclei issues.

2. **RC:** Other recent aerosol models with similar capabilities regarding the simulation of aerosol mixing state are described by Seland et al. (2008) and Wang et al. (2009).

   AR: We added references to Seland et al. (2008) and Wang et al. (2009) in the introduction.

3. **RC:** Other global models which attempt to simulate ice nuclei should be also mentioned: Liu et al. (2009); Gettelman et al. (2010); Salzmann et al. (2010); Hoose et al. (2010).

   AR: These models simulate the freezing processes itself, while MADE-in simulates the aerosol microphysics that concurs to the modification of ice nuclei properties. Having changed the focus of the paper, references to these models were not pertinent anymore.

4. **RC:** p 2227, l 7: "EMAC/MADE-IN keeps track of aerosol particles with different freezing ability" - this is overselling the model.

   AR: This sentence has been omitted and the capabilities of EMAC/MADE-in have been explained more in details.

5. **RC:** Please give the mode diameter ranges for the different modes and the diameters at emission. How is the interception diameter calculated?

   AR: The following parts have been added to the text:

   - Sect. 2.1.1: Similarly to MADE, the Aitken modes typically contain particles smaller than 100 nm and the accumulation modes have a typical size range of 100 nm to 1 μm. The size range of one mode is not fixed, and can change due to microphysical processes. The growth of particles, for instance, shifts the diameter of the modes toward larger values. The nucleation of many small particles shifts the mode to smaller diameters.
   - Sect. 2.3: The number of particles emitted in the Aitken mode is calculated assuming a number size distribution with median radius 0.015 μm and standard deviation 1.8. For deriving the number of particles emitted in the accumulation mode a median radius of 0.04 μm and a standard deviation of 1.8 are assumed, except for mineral dust and sea salt. The dust particle number emissions fluxes are considered as calculated by Dentener et al. (2006) taking into account variable size distributions. The emitted dust is assigned to the coarse mode (1664 Tg per year) and to the externally mixed accumulation mode acc$_{ext}$ (10.6 Tg per year), since the major emission regions of dust are usually poor of soluble material.
   - Sect. 2.1.2: The intersection diameter is calculated from the definition of log-normal mode, imposing that the number concentration of the Aitken and of the accumulation mode at D_N must be the same. More detailes about the technical aspect of the algorithm used can be found in Binkowski and Roselle (2003).

6. **RC:** equation (6): What exactly does the "aging" term refer to? (Other than coagulation and growth?)

   AR: The following has been added to the explanation of Eq. 6:

   $\frac{\partial N_i}{\partial t}\Big|_{\text{aging}}$ accounts for the number of particles that undergo aging due to condensation of sulfuric acid and/or organic vapor as well as uptake of water. The aging due to coagulation is included in $\frac{\partial N_i}{\partial t}\Big|_{\text{coag}}$. Mode merging (growth) takes place

only between modes of the same family, i.e. only between akn$_\text{sol}$ and acc$_\text{sol}$, between akn$_\text{ext}$ and acc$_\text{ext}$, and between akn$_\text{mix}$ and acc$_\text{mix}$.

7. **RC:** p 2234, l 6: Why is sea salt partitioned between the modes?

   AR: In the model set-up used in this work, sea salt is not included in the Aitken mode, therefore the sea salt concentration is zero. The model, however, allows for the presence of sea salt in the Aitken mode, too. The manuscript has been modified to make this clear.

8. **RC:** Condensation: As the Whitby et al. (1991) reference is not easily available, the calculation of the G_i terms should be explained in more detail. I understand that they depend on the second moment (the surface), but not why they should depend on the first moment (p 2235, l 1). And coming back to the point made above: the coarse mode would provide a relatively large surface. Why is this process excluded?

   AR: Descriptions of the calculation of the coagulation and condensation coefficients have been added as appendix. Binkowski and Roselle (2003) estimated the error related to the omission of the coarse mode using an average continental size distribution, showing that the interaction between the sub-micrometer modes and the coarse mode is weak. For a typical urban aerosol distribution Seinfeld and Pandis (2006) (Fig. 8.11) showed that the surface distribution is clearly dominated by the accumulation mode. We agree, however, that the coarse mode provides large surface areas, particularly close to its source regions. A better description of the coarse mode is planned for future versions of the model. Given our interest in the UTLS, we decided for the time being to focus on the sub-micrometer modes. Excluding the treatment of the coarse mode allow us to increase the details in the description of the Aitken and accumulation modes, while keeping the computational expenses low.

9. **RC:** p 2240, l 5: Is this referring to collision scavenging only?

   AR: Externally mixed BC and dust cannot be scavenged through nucleation scavenging in our model, but only through impact scavenging. This takes into account the probably low hygroscopicity of externally mixed BC and dust. We explain this point in Sect. 2.3.

10. **RC:** Is sulfate formation in the aqueous phase included in the model?

    AR: Sulfate formation in the aqueous phase is included through the following reactions, calculated by the submodel SCAV

    - $SO_3^{2-} + O_3 \rightarrow SO_4^{2-}$

    - $HSO_3^- + O_3 \rightarrow SO_4^{2-} + H^+$

    - $HSO_3^- + H_2O_2 \rightarrow SO_4^{2-} + H^+$

    We have extended the list of reactions in the supporting material to include also the liquid phase chemistry.

11. **RC:** equation (19): Please explain better what this equation represents.

    AR: We changed the explanation of Eq. 19 in the following way:

    The number concentration $N_\text{transferred}$ of soluble particles that is contaminated by Aitken mode BC is calculated from the number concentrations of the particles that are taken up by cloud droplets as

$$N_\text{transferred} = \min \left[\frac{N_{\text{acc}_\text{sol}}\Big|^\text{cl}}{N_{\text{acc}_\text{tot}}\Big|^\text{cl}} \left(N_{\text{akn}_\text{mix}}\Big|^\text{cl} + N_{\text{akn}_\text{ext}}\Big|^\text{cl} \right), N_{\text{acc}_\text{sol}}\Big|^\text{cl} \right], \quad (1)$$

where the number concentrations labelled with index "cl" refer to aerosol taken up by cloud particles. It is assumed here that only accumulation mode particles can be activated. $N_{acc_{tot}}\big|^{cl}$ is the total number of accumulation mode particles in cloud droplets. $N_{transferred}$ denotes the number of particles that is transferred from the soluble accumulation mode to the internally mixed modes containing BC. The Aitken mode BC particles merged with acc_{mix} and acc_{ext} within cloud particles do not induce transfer of particles between soluble and insoluble modes, because these accumulation modes already contain insoluble mass. However, they induce a transfer of mass from the Aitken modes containing BC to the internally mixed accumulation mode containing BC. If enough soluble accumulation mode particles are available, it is assumed that each particle in acc_{sol} merges with at most one Aitken mode BC particle. If more Aitken mode BC particles are scavenged than soluble accumulation mode particles, all soluble particles are transferred to the internally mixed accumulation mode with BC and $N_{transferred} = N_{acc_{sol}}\big|^{cl}$. Note that the Aitken mode does not contain mineral dust, owing to the comparatively large sizes of mineral dust particles.

12. **RC:** p 2242, l 12: Please specify how much dust is emitted into the coarse and accumulation modes, respectively.

    AR: We added the amount of dust emitted in each mode, which is 1664 Tg dust per year to the coarse mode and 10.6 Tg to the accumulation mode.

13. **RC:** Section 3: The only comparison with observation which refers to the simulated dust are the vertical profiles from the SAMUM campaign, one of which is not well simulated. As explained above, I don't consider this to be sufficient. More comparisons, as for example shown in the recent paper by Huneeus et al. (2010), should be included.

    AR: The profiles shown in Fig.9 evaluate the number concentration against SA-

MUM observations. We do not fully agree with the reviewer stating that one of the SAMUM profiles does not agree well. As explained in Sec.3.2, the background aerosol concentration, i.e. the one not perturbed by dust storms, is simulated very well. Specific dust storms cannot be represented in the model with the current set-up, since we used climatological emissions for dust. However, we have included a comparison between our results and dust mass observations as discussed by Huneeus et al. (2010), and between the total burdens of dust simulated by EMAC/MADE-in and Aerocom values (Textor et al., 2007).

14. **RC:** Is EMAC/MADE-IN also simulating AOD? If yes, this should be shown here.

    AR: EMAC/MADE-IN simulates also AOD. The aerosol optical properties are calculated for each individual mode through a linear combination of the properties of the different aerosol components, weighted with their respective volume fractions. We analyzed the AOD in dusty areas (not shown in the paper), and found that it is lower than the AOD observed by MODIS. We expected this result, given the comparatively low dust burden of the model. In this paper we aim at introducing the new model EMAC/MADE-in and to evaluate the prognostic variables of the aerosol module (mass and number concentrations of different modes). AOT is calculated by EMAC/MADE-in diagnostically from the aerosol mass concentrations. Therefore, we did not include any evaluation of AOT.

15. **RC:** Section 3.4: Please refer also to Lohmann and Hoose (2009), who attempted a similar comparison and similar sensitivity experiments.

    AR: The following text has been added to Sect. 3.4:

    These comparisons suggest that the efficiency of the aging process of insoluble particles as represented in the current version of the model may be comparatively high. In the global model studies by Bauer et al. (2008) and Lohmann and Hoose (2009) similar representations of BC aging as applied here were considered. Also these models tend to simulate higher fractions of internally mixed BC as

suggested by the observations. This reveals that the BC ageing as currently represented in these models may be too efficient. However, this conclusion should be carefully considered, given the uncertainties inherent in the measurements and possible systematic differences caused by limitations in the comparability of the model results and the observations. To asses possible uncertainties in the model representation of insoluble particle aging, further research is necessary applying also alternative methods to simulate the aging process.

16. **RC:** Please comment on the magnitude of BC emissions from aircraft and on how large the expected error is when they are omitted.

    AR: The following paragraph has been added to the manuscript: Hendricks et al. (2004) found by means of global simulations with a mass-based aerosol module that the amount of BC from aviation at 250 hPa (main flight level) could represent only a few percent of the total large-scale mean BC mass. The highest contributions were found over Europe and within the North Atlantic flight corridor. A corresponding estimate of the number contribution based on prescribed size distributions, suggests that BC particles from aircraft could contribute up to 30% to the total BC particle number concentration, noticeably increasing the number concentration of insoluble particles in the UTLS. Hendricks et al. (2004) considered emission data representative for the years around 1990. An update for these results by consideration of BC from aviation in EMAC/MADE-in could be a subject of future research.

17. **RC:** The supplementary material should be explained (or omitted).

    AR: We have included an explanation of the content of the supplementary material. We included the supplementary material for reasons of the reproducibility of our simulation, therefore it is mainly technical material needed to reproduce the set-up of new simulations with EMAC/MADE-IN.

18. **RC:** Table 5: Please include also the simulated burden of NO3 and NH4 (although

this is not available for the other models).

AR: The burdens of NO_3 and NH_4 have been included in Table 5.

Technical comments. We thank the reviewer for the technical comments, we have modified the text accordingly.

References

Bauer, S. E., Wright, D. L., Koch, D., Lewis, E. R., McGraw, R., Chang, L.-S., Schwartz, S. E., and Ruedy, R.: MATRIX (Multiconfiguration Aerosol TRacker of mIXing state): an aerosol microphysical module for global atmospheric models, Atmos. Chem. Phys., 8, 6003-6035, 2008.

Binkowski, F. and Roselle, S. J.: Models-3 Community Multiscale Air Quality (CMAQ) model aerosol component. 1. Model description, J. Geophys. Res., 108, 4183, 2003.

Cziczo, D. J., Froyd, K. D., Gallavardin, S. J., Moehler, O., Benz, S., Saathoff, H., and Murphy, D. M.: Deactivation of ice nuclei due to atmospherically relevant surface coatings, Environ. Res. Lett., 4, 044 013, 2009.

DeMott, P., Chen, Y., Kreidenweis, S. M., Rogers, D. C., and Sherman, D. E.: Ice formation by black carbon particles, Geophys. Res. Lett., 26, 2429-2432, 1999.

DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impact on climate, P. Natl. Acad. Sci. USA, 107, 11217-11222, 2010.

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., 6, 4321-4344, 2006.

Gettelman, A., Liu, X., Ghan, S. J., Morrison, H., Park, S., Conley, A. J., Klein, S. A., Boyle, J., Mitchell, D. L., and Li, J.-L. F.: Global simulations of ice nucleation and ice supersaturation with an improved cloud scheme in the Community Atmosphere Model, J. Geophys. Res., 115, D18 216, doi:10.1029/2009JD013797, 2010.

Hendricks, J., Kärcher, B., Döpelheuer, A., Feichter, J., Lohmann, U., and Baumgardner, D.: Simulating the global atmospheric black carbon cycle: a revisit to the contribution of aircraft emissions, Atmos. Chem. Phys., 4, 2521-2541, 2004.

Hoose, C., Kristjánsson, J. E., Chen, J.-P., and Hazra, A.: A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model, Journal of the Atmospheric Sciences, 67, 2483-2503, doi:10.1175/2010JAS3425.1, 2010.

Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Kinne, S., Prospero, J., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Fillmore, D., Ghan, S., Ginoux, P., Grini, A., Horowitz, L., Koch, D., Krol, M. C., Landing, W., Liu, X., Mahowald, N., Miller, R., Morcrette, J.-J., Myhre, G., Penner, J. E., Perlwitz, J., Stier, P., Takemura, T., , and Zender, C.: Global dust model intercomparison in AeroCom phase I, Atmos. Chem. Phys. Discuss., 10, 23781-23864, doi:10.5194/acpd-10-23781-2010, 2010.

Kärcher, B., Hendricks, J., and Lohmann, U.: Physically based parametrization of cirrus cloud formation for use in global atmospheric aerosol, J. Geophys. Res., 111, D01205, 2006.

Kärcher, B., Möhler, DeMott, Pechtl, and Yu: Insights into the rool of soot aerosols in cirrus cloud formation, Atmos. Chem. Phys., 7, 4203-4227, 2007.

Koehler, K. A., Kreidenweis, S. M., DeMott, P. J., Petters, M. D., Prenni, A. J., and Möhler, O.: Laboratory investigations of the impact of mineral dust aerosol on cold cloud formation, Atmos. Chem. Phys., 10, 11955-11968, 2010.

Liu, X., Penner, J. E., and Wang, M.: Influence of anthropogenic sulfate and black carbon on upper tropospheric clouds in the NCAR CAM3 model coupled to the IMPACT global aerosol model, J. Geophys. Res., 114, D03 204, 2009.

Lohmann, U. and Hoose, C.: Sensitivity studies of different aerosol indirect effects in mixed-phase clouds, Atmos. Chem. Phys., 9, 8917-8934, 2009.

Möhler, O., Buettner, S., Linke, C., Schnaiter, M., Saathoff, H., Stetzer, O., Wagner, R., Kraemer, M., Mangold, A., Ebert, V., and Schurath, U.: Effect of sulfuric acid coating on heterogeneous ice nucleation by soot aerosol particles., J. Geophys. Res., 110, D11 210, 2005.

Möhler, O., Benz, S., Saathoff, H., Schnaiter, M., Wagner, R., Schneider, J., Walter, S., Ebert, V., and Wagner, S.: The effect of organic coating on the heterogeneous ice nucleation effciency of mineral dust aerosols, Environ. Res. Lett., 3, 025 007, doi:10.1088/1748-9326/3/2/025007, 2008.

Salzmann, M., Ming, Y., Golaz, J.-C., Ginoux, P. A., Morrison, H., Gettelman, A., Krämer, M.,

C890

and Donner, L. J.: Two-moment bulk stratiform cloud microphysics in the GFDL AM3 GCM: description, evaluation, and sensitivity tests, Atmospheric Chemistry and Physics Discussions, 10, 6375-6446, doi:10.5194/acpd-10-6375-2010, 2010.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics., Wiley, 2nd edition, 2006.

Seland, Ø., Iversen, T., Kirkevåg, A., and Storelvmo, T.: Aerosol-climate interactions in the CAM-Oslo atmospheric GCM and investigation of associated basic shortcomings, Tellus, 60A, 459-491, 2008.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, T., Kloster, S., Koch, D., Kirkeväg, A., Kristjansson, J., Krol, M., Lauer, A., Lamarque, J., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: The effect of harmonized emissions on aerosol properties in global models - an AeroCom experiment, Atmos. Chem. Phys., 7, 4489-4501, 2007.

Wang, M., Penner, J. E., and Liu, X.: Coupled IMPACT aerosol and NCAR CAM3 model: Evaluation of predicted aerosol number and size distribution, J. Geophys. Res., 114, D06 302, 2009.

Whitby, E. R., McMurray, P., Shankar, U., and Binkowski, F.: Modal Aerosol Dynamics Modeling, Tech. Rep. 600/3-91/020, Atmospheric Research and Exposure Assess. Lab., U.S. Environmental Protection Agency, Research Triangle Park, available as NTIS PB91- 161729/AS, 1991.

C891