

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

Anonymous Referee #2

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General comments

Aquila et al. present an extension of the EMAC/MADE global aerosol-climate model. Their extension includes four new modes (two Aitken modes and two accumulation modes), such that externally mixed dust and BC, coated dust and BC and particles without insoluble core can be separated. The new model is evaluated against a number of surface and aircraft observations. I recognize the substantial amount of hard work which has gone into the model development. The model description is detailed and well written. The comparison to observations is done more carefully than for many other recent models (except that I am missing an evaluation of the dust component, see below). So far I find this manuscript of high quality and suitable for publication

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in GMD, but the authors are trying to stretch their results further and speak of the "simulation of potential IN". Here I don't follow the authors' line of reasoning for a number of reasons. My recommendation is to remove/amend parts of the manuscript referring to the simulation of ice nuclei. Many insights from recent laboratory and field studies on ice nuclei are ignored here.

My main point of criticism is that this model does not simulate IN. Thus the manuscript title and the model name are inaccurate. I question whether a more accurate simulation of the mixing state of Aitken and accumulation mode dust and BC particles is such a big step forward for simulation of ice nuclei concentrations in global models, as the authors claim. I ask the authors to consider the following points:

- There is contradictory evidence on whether coatings suppress ice nucleation, have no effect or even enhance it (papers to be cited here include Sullivan et al. (2010b,a)). According to some studies (e.g. Ansmann et al. (2008)), 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.
- 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.
- 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.
- "Potential IN" is a term invented by modellers and is not well defined. What measureable quantity does this correspond to? Does this refer to all particles which can induce heterogeneous above a certain temperature? If so, above what

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temperature? You should note that in no laboratory or 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.)

- DeMott et al. (2010) showed a remarkable correlation of IN with concentrations of aerosol particles larger than $0.5\mu\text{m}$. This points to a first-order dependence on size rather than on mixing state.
- 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.
- For the purpose of simulation of IN, I see a major problem in the fact that the “ acc_{ext} ” and “ acc_{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?

Detailed comments

- 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.
- 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).
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- 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).
- p 2227, l 7: “EMAC/MADE-IN keeps track of aerosol particles with different freezing ability” - this is overselling the model.
- Please give the mode diameter ranges for the different modes and the diameters at emission. How is the interception diameter calculated?
- equation (6): What exactly does the “aging” term refer to? (Other than coagulation and growth?)
- p 2234, l 6: Why is sea salt partitioned between the modes?
- 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?
- p 2240, l 5: Is this referring to collision scavenging only?
- Is sulfate formation in the aqueous phase included in the model?
- equation (19): Please explain better what this equation represents.
- p 2242, l 12: Please specify how much dust is emitted into the coarse and accumulation modes, respectively.
- 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.

- Is EMAC/MADE-IN also simulating AOD? If yes, this should be shown here.
- Section 3.4: Please refer also to Lohmann and Hoose (2009), who attempted a similar comparison and similar sensitivity experiments.
- Please comment on the magnitude of BC emissions from aircraft and on how large the expected error is when they are omitted.
- The supplementary material should be explained (or omitted).
- Table 5: Please include also the simulated burden of NO₃ and NH₄ (although this is not available for the other models).

Technical comments

- p 2245, l 1: (Spackman et al., 2010) is missing in the list of references.
- p 2256, l 15: 350 hPa → 300 hPa (?)
- p 2258, l 19: model → models
- p 2268, l 30: Higroscopic → Hygroscopic
- p 2269, l 7: Binkowsky → Binkowski

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