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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Sensitivity studies of different aerosol indirect effects in mixed-phase clouds" *by* U. Lohmann and C. Hoose

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**General Comment**

This study reports on a comprehensive set of simulations performed with a modified version of the ECHAM5 global model. It is enjoyable and relatively easy to read. The primary focus of the paper is on differences in indirect effects due to recent changes made to the physics of the model, including the sensitivity to the ice nucleation mode (especially) of black carbon (BC) and the incorporation of a revised version of the Bergeron-Findeisen (BF) process. Changes to the BF process are intended to make the simulations more realistic in the way that the water phase is partitioned, which can affect various feedbacks. For example, an increase in ice nuclei number concentrations may not lead to monotonic increases in ice crystal concentrations or precipitation



because the BF process limits new ice formation. If I understand correctly, the model with modifications is now less likely to glaciate, so can produce mixed phase and liquid clouds more readily. I can imagine that this means that the model will be more sensitive to specification of ice nucleation processes. Details of the realization of ice nucleation in the model compared to what exists in the atmosphere, including the activation spectra and number concentrations of ice nuclei available could lead to different results in the simulation of impacts of aerosols on mixed phase clouds. My primary suggestion is for this topic to be given some extra discussion based on the specific comments I list below. I realize that choices deemed reasonable have to be made of course in analyzing indirect effects within a global model.

The paper concludes with a call for more studies of ice nucleation, contact freezing nucleation in particular. I support this conclusion, but I think it is possible to overstate the need for contact freezing investigations, depending very much on the choices made for expressing contact nucleation by BC and the relatively weak strength of present evidence that IN coating clearly leads to a deactivation effect. Focusing on the first point only, there are two aspects that I feel need attention. First, contact freezing occurs for Aitken mode BC. Second, their potential numbers and freezing efficiencies are difficult for me to discern from the paper. Both Lohmann and Diehl (2006) and Hoose et al. (2008) are referenced regarding contact freezing by BC, each treating contact freezing by BC differently. It would be important to say if BC number concentrations that may freeze via contact freezing exceed present estimations of the numbers of contact freezing nuclei in air. If so, this should be made clear so that any new experiments can be finely focused on the assumptions made. Hoose et al. (2008) argue for the possibility that we may simply not have properly assessed the contact freezing activity of small externally mixed BC. My opinion is that this is highly unlikely. The activity ascribed to small BC in Hoose et al. (2008) suggest to me that small BC are nearly as efficient ice nuclei by contact freezing as reported some of the best AgI-based ice nuclei ever manufactured by combustion (DeMott 1995). While AgI-based IN may be quite different than other natural or anthropogenic IN, that study also demonstrated

the size dependence of contact freezing (smaller particles are the least efficient IN) that normally exists for any ice nucleation mechanism. Since Brownian diffusion is the primary scavenging mechanism in this size regime, Deshler and Vali (1992) should have detected the presence of high number concentrations of BC or any other species acting as contact freezing nuclei in their measurements. They did not. The number concentrations of contact freezing ice nuclei inferred in their study was around 1-10 per liter at -18°C, if the sizes of the colliding aerosols were assumed to be in the range of 0.02 to 0.1 micron diameter. Concerns may remain about the representativeness of previous contact freezing measurements, which the authors are welcome to state, but I note that the number concentrations of contact freezing nuclei measured by Deshler and Vali (1992) are similar to the numbers of ice nuclei we measure to be present in the atmosphere as condensation freezing/immersion freezing nuclei (Eidhammer et al. 2009). One might expect that the contact and immersion freezing ice nuclei numbers should be of the same order by number, since, just as is assumed in this model, they may be the same basic population of particles (just expressing their activity under different conditions). Thus, the numbers of BC allowed to act as freezing nuclei is a critical parameter to get correct in the model if one is to predict anthropogenic impacts of BC. I know that the authors understand this, just wish clarification. I struggled to find the information on the number concentrations of BC in air, since most figures here or in Hoose et al. (2008) list integrated tropospheric concentrations. I infer values of near 1 $cm^{-3}$ integrated; perhaps that is incorrect. It stands to reason that all of these particles should not be available to act as contact nuclei, just as laboratory and atmospheric studies show that only small fractions of various mineral dusts are available for freezing at warmer than -30°C (Field et al. 2006; Welti et al. 2009). Similarly, Diehl and Mitra (1998) found less than 1 in 10 soot particle collisions to freeze large drops, even as cold as -25°C. Can you confirm/list typical number concentrations and the concentrations available as potential contact freezing nuclei, and the specific references for the activation spectra (number fraction freezing versus temperature) for both BC and dust? I realize that you do not wish to repeat this information in every publication, but



it is critical information for understanding the role of inputs used as the model physics evolves in subsequent publications.

A critical concern that comes to mind is that if the activity and numbers of contact freezing BC have been grossly overestimated, it is not true that climate is as sensitive to anthropogenic ice indirect effects as has been previously inferred. On the other hand, it appears that there is no manner to alter the equivalent forcing by more than a few tenths of a W $m^{-2}$ in this model. Of course, once nucleation processes are refined, I can imagine that sensitivities might be better revealed.

Thus, my comments are focused less on the simulation results themselves and more on assuring that the detailed assumptions are clearly stated and evaluated as presently reasonable and supported.

**Specific Comments**

1) Introduction

P. 15048, lines 3-7: “If the glaciation effect prevails or not is very sensitive to the assumption of the ice nucleation abilities of natural mineral dust aerosols.” As I suggested above, whether or not the glaciation indirect effect prevails may as well be very sensitive to the assumption of ice nucleation abilities of black carbon particles; not only their efficiency for freezing by contact, but the number concentrations capable of doing so.

P. 15048, lines 19-27: As a question of clarification, were the aerosol concentration fields exactly the same between Lohmann and Diehl (2006) and the ECHAM5 studies; just the mixing state changed?

P. 15049, lines 3-5: “Thus, if in present times more dust aerosols are internally mixed immersion nuclei they are worse IN than in pre-industrial times, where more of them acted as contact nuclei.” Is it safe to assume that no coatings were possible in pre-industrial times. What about biogenic SOA?

P. 15049, line 9: Equating an alteration in mechanisms to a deactivation effect may

be misleading. It is, as you make clear, an assumed change in the ice nucleation efficiency due to a change in mechanism. In this regard, I would like to mention that the idea of altering mechanisms via a change in water uptake properties has in the past been exploited to manufacture ice nuclei that would act more (not less) efficiently because the slow contact freezing process was altered to a more rapid condensation freezing mechanism (Feng and Finnegan, 1984; DeMott, 1995). Specifically, the high ice nucleating efficiency of Ag-$I_x$-$Cl_y$ ice nuclei was retained by introducing a non-complexing hygroscopic material (NaCl). While it is possible that the particles produced by combustion in this case were not really coated in the sense one imagines happening in polluted atmospheres, it must raise a caution about the fact that we do not yet know for sure if coating represents an equivalent deactivation. Other subtleties not really discussed here include the fact that contact freezing is limited by collection times and the likely low numbers of IN capable of acting this way (which may or may not be well represented in the model at present – point already made above). In this scenario, it is critically important to be certain of the effective decrease in freezing temperature that the switch to condensation/immersion freezing leads to, another factor that may not be well known for many natural dust distributions (or BC).

P. 15049, lines 10-26: While it is a possibility, I think the present evidence for true deactivation by coatings in the heterogeneous regime warmer than 35°C is relatively modest. The Möhler et al. (2008) reference given here relates to impacts in the cirrus regime between 205 and 210K, so should probably be removed as supporting the case. The Eastwood et al. (2009) study shows a clear impact of coatings, but these impacts have thus far been measured only for kaolinite particles exceeding 5 microns in size that begin freezing at RH well below water saturation. Similar behavior has yet to be demonstrated for particles in the primary number mode of dust size distributions in the atmosphere. The author's group has recently shown that untreated kaolinite particles at sizes below 800 nm do not activate ice formation below water saturation at 245K (Welti et al., 2009). This has also been demonstrated by a number of other groups for additional natural dusts during a special workshop in 2007, as papers in a



forthcoming special issue of ACPD will show. Also relevant in this case may be the paper of Niedermeier et al. (2009). If water saturation and dilute droplet activation is required for dust freezing at these temperatures, one begins to wonder if a coating greatly matters. It is only clearly understood that a coating matters if there is a physiochemical transformation of the dust surface due to dissolution or chemical reaction or bonding, as you note mentioned in the Baker and Peter (2008) reference. In fairness, I note that the important issue of number concentrations of natural and anthropogenic IN is mentioned in the last sentence of this paragraph. This probably needs to be brought forward somehow.

2) Model Description

P. 15051, lines 10-11: Number distributions here and in Hoose et al (2008) are displayed as integrated tropospheric column values in units per square meter. I think it would help for readers to know the explicit volumetric number concentrations of aerosol, for example in a zonal mean snapshot. I may be calculating this wrong, but I think I infer column integrated numbers of >1 $cm^{-3}$ of potential contact nuclei. These are high numbers of ice nuclei based on any present measurements, except in perturbed situation (e.g., DeMott et al. 2003; Stith et al. 2009)

3) Set up of observations

I suggest adding a little more detail to Table 1 in regard to differences between the new simulations and Lohmann et al. (2008): Perhaps it should read something like, "Differences to Lohmann et al. (2008) are the incorporation of aerosol nucleation due to cosmic rays and organic vapors, a new water uptake scheme, thermophoresis for contact freezing, and addition of a size-dependent below cloud scavenging routine." I found it very hard to recall all of the differences that would have the impacts shown in the results figures.

4) Comparison with observations

Figure 4: There seems a problem with the vertical scale?

P. 15057, lines 15-20: The strong interplay between freezing and the BF process again stresses the need for repeating assumptions made about ice nucleating aerosols maximum concentrations) in this paper. It is hard to tell if the freezing at higher altitudes ends up dominated by homogeneous or heterogeneous freezing.

**Technical Corrections** P. 15053, lines 2-3: "Simultaneous growth of cloud droplets and ice crystals is not foreseen in our model." I assume this means it does not occur or is not possible. Please clarify this point.

P. 15057, line 14: Suggest replacing "this" with "the BF" to make the meaning of the sentence clearer regarding the different "frequencies" discussed.

**Additional References**

DeMott, P. J., Sassen, K., Poellot, M., Baumgardner, D., Rogers, D. C., Brooks, S., Prenni, A. J., and Kreidenweis, S. M.: African dust aerosols as atmospheric ice nuclei. Geophys. Res. Lett., 30, No. 14, 1732, doi:10.1029/2003GL017410, 2003.

DeMott, P.J.: Quantitative descriptions of ice formation mechanisms of silver iodide-type aerosols. Atmospheric Research, 38, 63-99, 1995.

Deshler, T., and Vali, G.: Atmospheric concentrations of submicron contact freezing nuclei, J. Atmos. Sci., 49, 773-784, 1992.

Diehl, K. and Mitra, S.: A laboratory study of the effects of a kerosene-burner exhaust on ice nucleation and the evaporation rate of ice crystals, Atmos. Environ., 32, 3145-3151, 1998.

Eidhammer, T., DeMott, P. J., and Kreidenweis, S. M.: A comparison of heterogeneous ice nucleation parameterizations using a parcel model framework. J. Geophys. Res., 114, D06202, doi:10.1029/2008JD011095, 2009.

Feng, D., and Finnegan, W.G.: An efficient, fast-functioning nucleating agent, AgI-



AgCl-4NaCl, J. Weather Modif., 21, 41, 1989.

Niedermeier, D., Hartmann, S., Shaw, R. A., Covert, D., Mentel, Th. F., Schneider, J., Poulain, L., Reitz, P., Spindler, C., Clauss, T., Kiselev, A., Hallbauer, E., Wex, H., Mildenberger, K., and Stratmann, F.: Heterogeneous freezing of droplets with immersed mineral dust particles – measurements and parameterization, Atmos. Chem. Phys. Discuss., 9, 15827–15865, 2009.

Stith, J. L., Ramanathan, V., Cooper, W. A., Roberts, G., DeMott, P. J., Carmichael, G., Hatch, C. D., Adhikary, B., Twohy, C. H., Rogers, D. C., Baumgardner, D., Prenni, A. J., Campos, T., Gao, R. S., Anderson, J., and Feng, Y.: An overview of aircraft observations from the Pacific Dust Experiment campaign. J. Geophys. Res., 114, D05207, doi:10.1029/2008JD010924, 2009

Welti, A, F. Lüönd, F., Stetzer, O., and Lohmann, U., Influence of particle size on the ice nucleating ability of mineral dusts, Atmos. Chem. Phys. Discuss., 9, 6929–6955, 2009.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15045, 2009.