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Comment

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

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Thank you, Paul, for your valuable comments and suggestions. The responses to your comments are marked in italics.

General Comment

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.

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This is a good point and something we started to think about lately as well. You are right that the contact freezing data for BC are rather disputable. Therefore we decided to repeat all simulations without contact freezing of BC. The conclusions remain rather similar though

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.

That's true. We added that

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?

No, they were not. In Lohmann and Diehl (2006), we also used older aerosol emissions. We added that

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 preindustrial times. What about biogenic SOA?

The de-activation effect assumes that coatings are dominated by anthropogenic aerosols, such as sulfates and nitrates, which increased since pre-industrial times. If,

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on the other hand, coatings were dominated by biogenic secondary organic aerosols (SOA) and coatings by anthropogenic species were negligible, the de-activation effect would be negligible. We added that

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-Ix-Cly ice nuclei was retained by introducing a noncomplexing 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).

These are good cautionary notes. We added them

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

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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 physio-chemical 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.

Thanks for these cautionary notes. We added them. We didn't move the number concentrations forward as we like them as a summary statement but mentioned number concentrations above in response to your first comment

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 $>1 \text{ cm}^{-3}$ 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)

Unfortunately we did not save the number of potential IN. Thus, this information is not available. Given that we are now omitting the contact freezing of BC, we hope that's ok. Note also that the column burdens of $N_{cnt,BC}$ in Hoose et al (2008) do not refer to

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particles which actually act as ice nuclei, but to BC particles which are externally mixed and therefore can theoretically contribute to the contact freezing process. A "contact IN" concentration is difficult to define with the ice nucleation parameterization used in ECHAM5-HAM. The parameterization goes back to Diehl and Wurzler (2004) and does not provide ice nuclei concentrations, but relates the freezing rate to the ambient aerosol composition. We realize that this can lead to unrealistically high freezing rates in situations with low aerosol load, but a high BC fraction, because no upper limit is applied to the number of BC particles acting as contact nuclei - thus our decision to omit contact freezing by BC.

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.

We added that

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

Sorry, the vertical axis was given in model levels. We corrected it to hPa

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.

We added contour lines of 273 and 238 K

Technical Corrections P. 15053, lines 2-3: "Simultaneous growth of cloud droplets and

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ice crystals is not foreseen in our model.” I assume this means it does not occur or is not possible. Please clarify this point.

It means that it is not possible. We changed that

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

Done

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