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Interactive comment on "The effects of hygroscopicity of fossil fuel combustion aerosols on mixed-phase clouds" *by* Y. Yun et al.

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General comments

Yun et al have included a new treatment of black carbon in the CAM-IMPACT GCM, which distinguishes hydrophobic, hydrophilic and hygroscopic particles by the existence and thickness of the sulfuric acid coating on these particles. Different ice nucleation abilities in the mixed-phase temperature range are assigned to these particles, and the resultant aerosol indirect forcing is calculated with the help of an offline radiation model from simulations for PD and PI aerosol emissions.

The authors have addressed my critical remarks on a previous version of this text by including more discussion on other ice nucleation experiments with soot particles, and by clarifying a misunderstanding about the importance of contact nucleation in their

model. These are certainly significant improvements. However, I still think that the ice nucleation parameterization used in this study and the ad-hoc modification applied to it are not suitable for the current purpose.

The below points are fundamental issues, which can only be addressed by thoroughly revising the treatment of ice nucleation in this model. I strongly encourage the authors to do so. The new treatment of hydrophobic, hydrophilic and hygroscopic soot is a very promising and novel development, which might even have other useful applications.

Major comments

- The parameterization by Phillips et al. (2008) has been critized earlier for allowing too much ice nucleation on soot at high subzero temperatures. This could lead to an overestimation of the contribution to soot to mixed-phase cloud glaciation in the base case. It should be shown and discussed how large this contribution is in the CAM-IMPACT model. The anthropogenic forcing for both the 1BC and the 3BC-version of the model critically depends on this. A revised version of the Phillips et al. (2008) parameterization is now available (Phillips et al., 2012) in which immersion freezing by soot is suppressed at the warmest temperatures.
- Using the results of Koehler et al. (2009) for the mixed-phase temperature range, i.e. above -38C and at water saturation (as far as I understand, water saturation is prescribed in the model in mixed-phase clouds) is an unsupported extrapolation of the measurements. Koehler et al. (2009) studied deposition and condensation nucleation in the cirrus temperature regime by increasing RH at a given temperature in their instrument. In mixed-phase clouds, however, most air parcels reach water saturation at some higher temperature (lower altitude) and then cool further while ascending, maintaining water saturation (Wiacek and Peter, 2009). This means that the inhibited condensation on hydrophobic soot, which Koehler

et al. (2009) also measured, might be irrelevant when the (immersion) freezing temperature is reached, as long as the particles have entered the droplets either due to higher peak supersaturations at cloud base or due to collision scavenging.

- In addition, taking the activated fractions at RHw=100% is dangerous because (as also discussed by Koehler et al. (2009)) CFDCs resolve RH poorly in the supersaturated regime (several percent uncertainty). In Petters et al. (2009), it was shown that the activated fraction for biomass burning particles at -30C increases at water supersaturation until a plateau was reached at about 9% supersaturation. I would expect a similar behaviour for soot. At RHw=100% and -40C, one likely measures only a (small) contribution from deposition nucleation and, if some fraction of the soot activates as CCN, the same fraction is expected to freeze homogeneously (-40C is already relatively far below the homogeneous freezing temperature). Both are not relevant for mixed-phase clouds.

- While the effect of sulfate coatings on soot ice nucleation properties in the mixed-phase temperature regime is highly uncertain, because essentially no measurements are available for these conditions, there are several studies (cited above) investigating these effects at cirrus conditions. In my opinion this would be a more useful application of the newly developed aerosol model. However, it appears that the IN parameterization is kept unchanged for cirrus conditions. How is this done without introducing inconsistencies?

Minor comments

- The new aerosol scheme deserves more discussion in the results section. In particular, comparisons to observations would be appropriate, e.g. to the number fraction of internally mixed BC particles measured by Schwarz et al. (2008). Also

some information on the BC burden in the model and how this is changed by the new treatment should be added.

- The ice nucleation parameterization, in particular if a number of changes are applied, should be shown graphically (e.g. as activated fraction for a given particle size as a function of RHw and T).

- Koehler et al. (2009) studied five different soots, three of which are selected for this study. Why not the other two? This seems arbitrary. (On a side-note, it took me a long time to figure out which data were actually used. This should have been stated more explicitly.)

- The authors state that the Bergeron-Findeisen process is included in the offline simulations. It is unclear to me how this is done. Glaciation of clouds via the BF-process can lead to precipitation and cloud dissipation, but how can this information be kept in the following timesteps in an offline model?

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