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

In this manuscript, a new parameterization scheme of ice nucleation was proposed and implemented in the CAM-IMPACT model, which was then applied to study the influence of elevated emissions on anthropogenic forcing. Sensitivity studies were also carried through comparison among different schemes). The parameterization of ice nucleation and mixed phase clouds is an important and challenging issue in climate modeling. I appreciate the authors' scientific work and I find their results interesting and promising. My major concern is if the new parameterization scheme makes more sense compared with the empirical equations. It seems that the provided experimental results are not sufficient to significantly improve the scheme (see general comments 1). Overall, I find the manuscript meets the scope of the journal and I would recommend publication of this work after the authors have seriously considered my comments/suggestions.

(Original texts in *italics*, my comments in plain font)

General comments

1. Ice nucleation activity of BC particles.

In the 3-ffBC/OM scheme, ice nucleation activity (INA) is considered as a function of the coating thickness of sulfuric acid. This assumption is based on (1) INA measurements of soot particles with different hygroscopicity and (2) the dependence of soot hygroscopicity on the coating thickness of sulfuric acid.

Besides sulfate, secondary organic matter, which comprises a major fraction of aerosol particle mass (Zhang, Jimenez et al. 2007), can also contribute to the coating and the change of aerosol hygroscopicity (Gunthe, Rose et al. 2011) and INA (Crawford, Möhler et al. 2011, as mentioned by the authors). The effects of organics could be as important as sulfate for both real atmosphere and laboratory studies. I am not sure if the scheme can be really improved by considering the coating of sulfuric acid alone. It seems that there is no comparable (to sulfuric acid) study about the effect of organics on INA. Therefore, I encourage the authors to provide complimentary information about all IN (ice nuclei, 3 kinds of BC, dust) under different schemes (1BC, 3BC_SCO and 3BC_noSCO), e.g. something like Fig. C1 (Phillips, DeMott et al. 2008) and Fig. C2 (Hoose, Kristjánsson et al. 2010). By adding such information, the readers could judge the scheme and conclusions themselves.

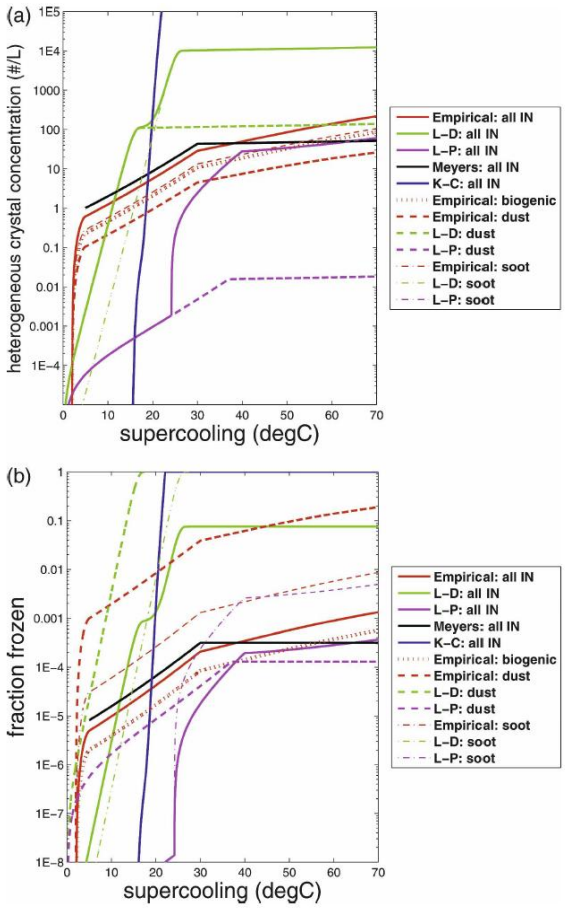

Figure C1: (a) Number concentration of heterogeneous crystals and (b) their freezing fraction for the three species of insoluble aerosol (adopted from Fig. 10 of Phillips et al, 2008).

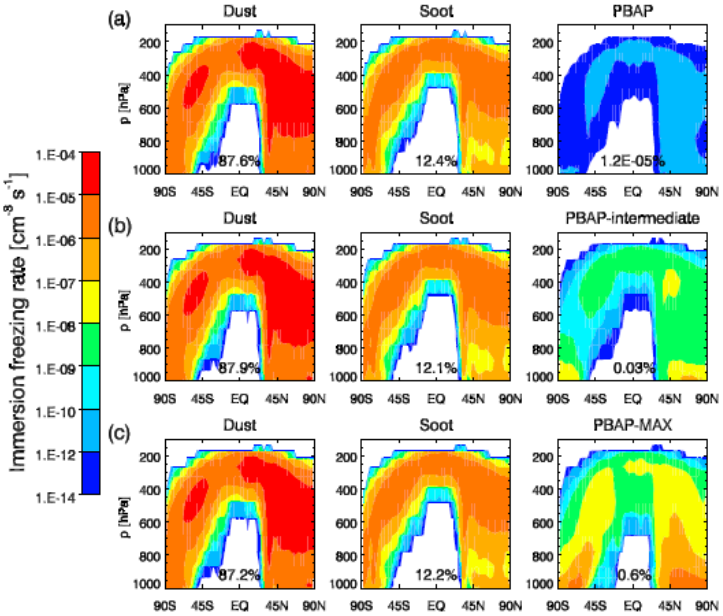

Figure C2: Zonal annual mean immersion freezing rates (a) in simulation PBAP, (b) in simulation PBAP-intermediate, and (c) in simulation PBAP-MAX (adopted from Fig. 4 of Hoose et al., 2010).

2. Mixing state of BC.

The mixing state of BC is a crucial parameter for its CCN, IN activity and optical properties. It is not explicitly explained how the mixing state of BC is treated in this study. Do you consider BC as externally mixed or internally mixed with other component (SO_4^{2-}, organics)? The authors state that BC and OM are assumed to be internally mixed (P19991 line 17, 'Although BC and OM are treated as distinct species in the model, they are assumed to be internally mixed'), how about BC and sulfate? The use of coating thickness (n_{coat}) seems to imply that the transition from externally mixed to internally mixed BC has been considered but I didn't find any details in this paper.

In the new scheme, hygroscopic BC is defined as BC particles with $n_{coat}>3$. Is there any upper limit of n_{coat} confining hygroscopic BC? Field measurements have found internally mixed particles with more coating materials (sulfate, organics) than a small BC cores, which is actually often the case in remote areas or even polluted areas (Cheng, Su et al. 2012). INA of such particles (thickly coated BC) is more closer to the coating material sulfate/organics. If there is no upper limit of n_{coat}, even a small BC core can make the whole particles to be BC, which has subsequent impact on the concentration of ice nuclei and mixed-phase clouds.

3. Hygroscopicity or IN activity

It might not be appropriate to use the current title considering the scope of this paper. It is more about ice nucleation ability rather than hygroscopicity. According to the current title, I would expect impact on the CCN activation, hygroscopic growth and deposition/life time of BC particles.

'The effects of hygroscopicity of fossil fuel combustion aerosols on mixed-phase clouds' changed to 'The effects of ice nucleation ability of fossil fuel combustion aerosols on mixed-phase clouds'

Specific comments

1. Abstract: Page 19988, line 7; Table 1

'The new scheme results in significant changes …'

Please specify which scheme (3BC_SCO or 3BC_noSCO) you meant. Also in Table 1, 3BC SCO scheme is described as *'3-ffBC/OM scheme and hygroscopic particles as heterogeneous ice nuclei'*, do you mean hygroscopic soot particles as

heterogeneous ice nuclei?

2. Abstract: Page 19988, line 10

'but could be more positive if hygroscopic soot particles are allowed to nucleate ice particles.'

I suggest the authors to give exact numbers for the case when hygroscopic soot particles are allowed to nucleate ice, 'but could be more positive (x W m^{-2} and y W m^{-2}) …'

3. Introduction: Page 19988, line 15

'Soot aerosols produced by fossil fuel and biomass burning contain both black carbon (BC) and organic matter (OM)…'

It is not appropriate to state that soot particles contain black carbon and organic matter, both of 'which act to absorb solar radiation'. Black carbon could contain compounds (grey carbon) which don't belong to soot particles. Also many/or probably most organic matters in the atmosphere is not actively involved in the absorption of solar radiation. I suggest the author to reformulate these claims and refer to the paper of (Andreae and Gelencsér 2006).

4. Page 19989, line 3

'During the lifetime of soot aerosols in the atmosphere their hygroscopicity can be altered through coating by sulfate (Zhang et al., 2008).'

Besides sulfate, hygroscopicity can also be modified through coating by organics (Petters and Kreidenweis 2007; Gunthe, Rose et al. 2011).

5. Page 19990, line 18

'Popovicheva et al. (2008, 2010) suggested that the hygroscopicity of soot could be quantified by the amount of water film extended over the soot surface at relative hu20 midity < 80%.'

Is the difference in hygroscopicity sufficient to explain the contrasting/different results of previous experiments? Considering the parameterization scheme, is there any other factor that we should include for future studies?

6. Page 19991, Line 8

'The coupled model (inline simulation) provides …'

Do you mean ‘online simulation’?

7. Page 19991, Line 8,

‘The coupled model (inline simulation) provides the aerosol fields and meteorology fields for the offline model, and is used to calculate the total anthropogenic forcing.’

I am wondering why both offline and online simulations are used. Could you please give some explanations?

8. Page 19991, Sect 2.1

‘The University of Michigan IMPACT aerosol model … a two-moment microphysics scheme … The PH08 parameterization was modified …

It is good scientific practice to briefly describe the method first and then complement it by references. It would be helpful to include brief introduction about the aerosol model, mocrophysics, etc.

9. Page 19991, Line 10

‘The offline model reads the aerosol and meteorology fields and examines the cloud water fields and mixed-phase cloud anthropogenic forcing without involving feedbacks to the cloud fields from changes in aerosols.’

It is not clear to me how the offline model works. Since hydrometers (both liquid and frozen water) generally belong to meteorology fields, ‘reading meteorology fields’ for me means that the model has got all information about mixed-clouds through input meteorological data. How aerosols change the prescribed meteorological fields in the offline model?

10. Page 19992, line 16

‘The accommodation coefficient for the,’

Remove the comma

11. Page 19992, Sect 2.2

I suggest the authors to express the new scheme by a series of equations. It is difficult for me to figure out how the new scheme works exactly.

12. P19993, line 13

‘Therefore, we made two assumptions to treat the freezing of hygroscopic particles at

higher temperatures. The first is … The second is that they freeze heterogeneously, and we scale the frozen fraction of PH08 by a factor of 15.'

Could you please specify the conditions when they are assumed to freeze homo- or hetero-geneously? Otherwise these assumptions seem to be contradictory.

13. Page 19994, line 13-17

'Hydrophobic ffBC/OM is confined… The lifetime of the ffBC/OM particles increases with hygroscopicity from 0.45 to 0.95 to 4.55 days.'

Do you consider the impact of soot hygroscopicity on its lifetime/removal in your model simulations? If so, please add some descriptions. If not, difference in lifetime should be the cause and difference in hygroscopicity the effect. In the current manuscript, it is expressed the other way round.

14. Page 19995, line 4,16

'Figure 3 shows the grid-mean ice number concentration (Ni) change from PI to PD' … 'An increase in Ni causes a net conversion of liquid to ice'

Is Ni the number concentration of ice nuclei or the hydrometer - ice? *'An increase in Ni causes a conversion of liquid to ice'* seems to imply Ni is the ice nuclei concentration. I suggest the authors to clarify this.

15. Page 20002, Table 2

If *'off-line forcings are for mixed-phase clouds'* only, should the forcings of Net Whole-Sky be left blank?

16. Page 20003, Fig. 1

The resolution of the figure is too low.

17. Page 20003, Fig.1, 2, etc

I suggest using the same format for all units, i.e. using either mg/m2 or mg m-2, not both. See figure titles and captions.

Reference:

Andreae, M. O. and A. Gelencsér (2006). "Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols." Atmos. Chem. Phys. **6**(10): 3131-3148.

Cheng, Y. F., H. Su, et al. (2012). "Size-resolved measurement of the mixing state of soot in the megacity Beijing, China: diurnal cycle, aging and parameterization." Atmos. Chem. Phys. **12**(10): 4477-4491.

Crawford, I., O. Möhler, et al. (2011). "Studies of propane flame soot acting as heterogeneous ice nuclei in conjunction with single particle soot photometer measurements." Atmos. Chem. Phys. **11**(18): 9549-9561.

Gunthe, S. S., D. Rose, et al. (2011). "Cloud condensation nuclei (CCN) from fresh and aged air pollution in the megacity region of Beijing." ATMOSPHERIC CHEMISTRY AND PHYSICS **11**(21): 11023-11039.

Hoose, C., J. E. Kristjánsson, et al. (2010). "How important is biological ice nucleation in clouds on a global scale?" Environmental Research Letters **5**(2): 024009.

Petters, M. D. and S. M. Kreidenweis (2007). "A single parameter representation of hygroscopic growth and cloud condensation nucleus activity." Atmos. Chem. Phys. **7**(8): 1961-1971.

Phillips, V. T. J., P. J. DeMott, et al. (2008). "An Empirical Parameterization of Heterogeneous Ice Nucleation for Multiple Chemical Species of Aerosol." Journal of the Atmospheric Sciences **65**(9): 2757-2783.

Zhang, Q., J. L. Jimenez, et al. (2007). "Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes." Geophys. Res. Lett. **34**(13): L13801.