Review of "Radiative forcing of anthropogenic aerosols on cirrus clouds using a hybrid ice nucleation scheme"

Summary:

The manuscript uses a newly developed ice nucleation scheme to examine the anthropogenic aerosol effect on cirrus clouds between preindustrial and present day conditions. The anthropogenic emissions have two opposite effects on the frequency of homogeneous ice nucleation and ice crystal number: soot particles decrease the homogeneous freezing and ice crystal number, while sulphur emissions enhance homogeneous nucleation. The total aerosol radiative effect is, moreover, strongly dependent on the freezing assumptions of the background state, as shown by the example of secondary organic aerosols. This manuscript has the potential to reveal some more details about the very uncertain anthropogenic aerosol effects on cirrus clouds. However, I am worried that the authors often describe features, which are not statistically significant. On one hand, they tried to avoid such feedbacks with limited success as the meteorological responses seem to often dominate over the cirrus microphysical responses to aerosols. I am therefore asking the authors to reassess their results after applying some form of a significance test. Adding several ensemble runs for each of their cases may help increasing the confidence in the presented results too. If those meteorological feedbacks turn out to still be important, than I would like to hear about them and understand them. This, together with my other comments, will demand substantial revision to the manuscript.

Major comments:

1.)
I fear many of the conclusions of the paper at the regional level are not robust. The authors may be simply describing climatic noise and not climatic responses of cirrus cloud microphysics to changes in anthropogenic emissions, particularly what described on pages 15-19. In particular, I do not understand why the main signal in most of the panels in Figures 5,6,7,9,10,14 has the most pronounced anomaly between SE Asia and the W Pacific. What is causing that? We need to know it better, if that feedback is real/statistically significant? Would the same occur also in a longer (at least 10 years) free-running simulation? Considering the small changes due to the anthropogenic forcing, it may be valuable to run several ensembles for each of the cases.
Why is the meterological response so dominant if you are nudging your result?
On the other hand, why not allowing the full extent of the dynamical responses? Ultimately, despite doing the nudging, the additional responses seem to often dominate over the pure aerosol signal.
It may also be beneficial to divide the radiative signal into the clear-sky and cloudy-sky (cloud radiative effect) components.

2.)
The authors provide a few numbers of the estimated anthropogenic aerosol radiative forcing. However, the model used may allow you to further experiment with preindustrial (PI) and present day (PD) aerosol burdens/emissions in order to get a range of possible radiative forcings, given the large range of uncertainty and the inability to directly verify models with some form of observational data on the homogenous vs. heterogeneous freezing issue. Could you, for example, engineer your PI climate to produce an extreme low heterogeneous nucleation scenario and an extreme high heterogeneous nucleation scenario and apply at this point the anthropogenic aerosol forcing. You could also have two setups with the lowest and the highest plausible ice nucleating ability of soot.
To finish this comment on a positive note: I think your SOA experiment is a good example of some of those sensitivity experiments that I can foresee and would give us a better range of aerosol effects.

3.)
The abstract reads as if a large part of the paper would be dedicated to a description of the newly developed scheme. However, this is a paper, which is using the scheme to estimate the aerosol effects on cirrus. You could include a more detailed description of your new freezing scheme.
It would be useful to have a more quantitative statement than “the ice number concentrations are in reasonable agreement/somewhat overestimated”. Was there an improvement compared with the previous nucleation schemes used by the same group?

I am also missing in particular a description of what aerosols species can nucleate ice heterogeneously and under what assumption. I understand the focus is on the soot, but the background loading of dust may have a huge impact on the magnitude of anthropogenic forcing. A sensitivity experiment with higher and another one with lower dust emissions may help addressing this issue.

4.)
What is your definition of a cirrus cloud? Is the paper referring only to the anthropogenic forcing on clouds at temperatures colder than the homogeneous freezing of water?

Is the estimated forcing including the direct radiative forcing by increased aerosol burden? If so, please distinguish the clear sky effects from changes in cloud radiative effects.

5.)
How does your model treat with cirrus with a origin at temperatures warmer than the homogeneous freezing threshold of water, also named “liquid-origin cirrus” (e.g. Krämer et al., 2016).
In particular, how are detrained ice crystals treated by the model? I understand the liquid-origin cirrus probably cannot be affected by aerosols, but the relative importance of detrainment vs. in-situ nucleation will

substantially limit the potential for anthropogenic aerosol forcing in regions with frequent deep convection.

6.)
Please use a reasonable number of significant digits when providing results. Adding the third significant number likely makes no sense with only 6 years of simulations (e.g. page 7, line 163 and 164 and so on…).

7.)
Some of the panels/figures need to be moved to the supplementary material due to the large amount of figures.
I also suggest that the vertical cross section plots are cut at about 700 hPa. Your focus is on cirrus clouds, while most of the plot area is wasted in the lower troposphere! (Fig. 3,4,5,6,13,14,15,16)

Specific comments:

page 1, lines 17-19:
please be more quantitative

page 1, line 23 also lines 25 and 34:
The number of Ni doesn't mean much to the large majority of readers. Relative anomalies in units of % change may be more appropriate. Moreover, why do you focus on Ni only? A change in ice crystal radius will also contribute to changes in cirrus lifetime, leading to changes in cirrus optical properties.

page 2, lines 29-30:
Does this include both clear (direct radiative forcing) and cloudy sky changes? Please mention both.

page 2, line 47:
Considering that you are talking about changes to cirrus radiative effects, a reference like Hong et al., 2016 and/or Matus et al., 2017 may be useful.

page 12, lines 283-285:
Could you also provide an effective or volumetric ice crystal radius histogram?

page 12, lines 305-307:
be more quantitative!

page 13, lines 313-316:
What about dust? Shouldn't dust be the dominant source of ice nucleating particles at cirrus levels globally?

page 13, lines 328-330:
I do not understand why the changes are largest at about 200 hPa in a region, that should be dominated by deep convective detrainment, therefore coinciding with peak detrainment level. Is your model simulating a too low

convective cloud top?
Alternatively, I could imagine that changes in homogeneous vs. heterogeneous freezing may not be as radiatively important in case the number of detrained ice crystals and ice water content dominates over the in-situ ice nucleation.

Why is the effect not larger in the midlatitudes, where soot emissions are the largest?

page 14, lines 353-354:
Direct effect vs. adjustments!

page 15, lines 366:
Why is there still a feedback on climatological state? Couldn't you use a longer simulation or nudge harder. A longer free-running simulation may tell something about the origin of those cloud adjustments, while a stronger nudging may prevent some of the noise to occur at first place.

page 15, lines 366- 394:
I am missing explanations that go beyond the "aerosol and cloud feedbacks to the meteorological state". Please describe what is really going on! Are the described patterns "real" at all? I am afraid that a lot of what mentioned is climatic or meteorologic "noise". Please apply a measure of statistical significance!
The meteorological feedbacks can be studied in an additional free running experiment. If such feedbacks are relatively speaking comparably or more important than the direct changes to cirrus freezing, we need to know more about them!

Chapter 3.2:
I am a bit skeptical about the explanations of causes leading to changes in IWP, LWP, and radiative fluxes. How can you be sure you are seeing more than simple climatic variability? Adding a form of significance would be a first and easy step that would help clarifying this issue.

Moreover, if the meteorological adjustments play a larger role than the changes to cirrus clouds alone, you should dedicate part of your manuscript to those adjustments and try to understand them. In free-running experiments.

Please distinguish between changes to clear-sky and cloud radiative effects due to changes in emissions/freezing. Total net SW/LW/net radiative fluxes represent a mix of direct aerosol radiative effects and their impact on cirrus (and maybe also other cloud through meteorological feedbacks). Please show changes in clear sky and CRE separately!

Moreover, it may be useful to separate the radiative perturbations on cirrus clouds only from the rest of the clouds. You could diagnose the cirrus cloud CRE with a help of a double call to the radiation routine, similarly to what is done for clear-sky radiative effects.

page 18, lines 451-452:
Why is the FNT from soot largest around 30°N? Aren’t the emissions much larger further north in the midlatitudes, around 40-60°N?

Fig. 3:
Why don’t we see heterogeneous freezing that originates from the main dust sources (Sahara, Taklimakan, maybe Australia) in the panel e ? Is dust allowed to act as an ice nucleating particle?
Panels a,c,e miss units!

Fig. 5,6,7,14,15,16:
Panels a,c,e miss units!

Fig. 10,11,12,17 also miss units!

Fig 10,11,12 and the corresponding text:

Please use a more descriptive naming for the FSNT,FLNT,FNT fluxes. Those abbreviation are not very intuitive to people outside of the CAM/CESM modeling community.

Reference:

- Hong and Liu, 2015: The Characteristics of Ice Cloud Properties Derived from CloudSat and CALIPSO Measurements, *JClim*
- Krämer et al., 2016: A microphysical guide to cirrus clouds – Part 1: cirrus types, *ACP*
- Matus and L’Ecuyer, 2017: The role of cloud phase in Earth's radiation budget, *JGR-A*