Review of Visioni et al.

This is a valuable contribution to the underrepresented topic of cirrus responses to stratospheric sulphur injections. While I think the authors did a good job in explaining the main physical mechanism behind the observed changes, I am pointing out a few more issues, which would need to be addressed before the paper can be published in final form.

Major comments

1.) Is detrained moisture/ice water content from convection included in the cirrus formation mechanisms? How did you include it?

You mention on page 7 that upper tropospheric ice can be formed only by homogeneous or heterogeneous freezing. However, a large part of the cirrus, in particularly in the tropics, is formed by detrainment of ice crystals from deep convective cores. Such ice crystals formed either by homogeneous nucleation of cloud droplets or in mixed phase by heterogeneous nucleation; their formation is therefore significantly different from the in-situ cirrus.

Did you include such detrained ice crystal sources in your model? I think the strength and level of maximum detrainment is probably modulating the responses of in-situ formed cirrus in the tropics, i.e. in the region where most of your cirrus cloud radiative effect comes from.

Most of your ice mass comes from heterogeneous freezing at lower elevations in the tropics. This is in a zonal average perspective not realistic, as most of it should be a result of detrainment from deep convection, at least near the location of the intertropical convergence zone.

Detrainment from deep convective clouds is an important source of cirrus clouds and therefore needs to be mentioned/commented in the manuscript.

2.) Model evaluation with MERRA2/MODIS data

I. Please state which version of MODIS data you use. You cite Yang et al., 2007, which is a reference for the V5. I assume you either use V5 or V6, please add this as the retrievals changed between several product versions. Do you use level 3 1x1° gridded data?

II. I would suggest removing the use of MODIS IC radius due to the following reasons:

• MODIS derived IC radius is valid only for cloud tops of optically thicker clouds and not representative of the whole cloud distribution. In a thick cloud, the MODIS IC effective radius would correspond to the upper

portion of the thick(er) cloud, until the optical depths of about 1.2, at least for the case of detrained anvil clouds as shown in Hong et al., 2012. The retrieval would give more weight to the radius closer to cloud top also for the case of intermediately thick cirrus (COD between 1 and 5, Zhang et al., 2010).

- MODIS cannot see the thinnest of the cirrus clouds. Its approximate detection limit is close to COD of 0.4 (Ackerman et al., 2008). I assume you include clouds of any optical depth in your analysis.
- MODIS is a passive instrument and detects cloud properties only during daytime, while I assume you take both day and night data from the model output

To summarize my point, the comparison of IC radius and the derived IC number concentration is based on too many very shaky assumptions and needs to be removed from the manuscript. If you would like to keep it, you may use the MODIS satellite simulator, which takes into account MODIS retrieval limitations and therefore ensures an apple-to-apple comparison.

III. <u>MERRA2</u>

MERRA2 has a very simplistic treatment of ice clouds, leading to large biases (e.g large biases in cloud radiative effect noted in Bosilovich et al., 2015). Using a reanalysis dataset is anyway not the best, but if you already went for one, ERA5 would be a more appropriate choice, as it compares better with CALIPSO-CloudSat datasets (DARDAR, 2C-ICE) as shown for instance by Duncan and Eriksson, 2018. Nevertheless, considering this is the second phase of review, I can accept the comparison used in Figure 1 as good enough due to large IWC retrieval uncertainty (as you also pointed out in the manuscript).

Yet, I think you should remove from the paper your optical depth estimates from MERRA+MODIS in figure 2, as the assumptions behind that plot are too large and you are mixing up reanalysis, satellite retrieval, and model output without making sure this is an "apple-to-apple" comparison (i.e. you don't take into account the satellite retrieval limitations and the issue of collocation of data in space and time).

Minor comments

• please add uncertainty estimates (e.g. +/- 1 st. dev.) to the results you show, at least in the tables. This would give the reader a better feeling for the significance of your radiative forcing anomalies.

Abstract:

After line 15 the abstract clarity becomes challenging for the reader as you are making very fast transitions from effects of cirrus clouds which cool the climate, to comparing all-sky with clear-sky forcing, and saying that the all-sky has a

positive effect on the radiative balance. I would just qualitatively mention the effect of a positive (total) cloud radiative effect -> dimming the sun that reaches the cloud tops indeed has to decrease the amount of reflected SW radiation. Moreover, do you really need to always mention 2 significant numbers after the decimal point, considering all the uncertainties?

page 2, line 21-24:

The current best knowledge of cirrus microphysics does not show much support of the predominance of homogeneous nucleation in in-situ cirrus cloud. Your extensive answer to reviewer #1 unfortunately does not help in changing that view. I think the uncertainty in cirrus formation mechanisms is high enough to accept your modelling results related to the freezing mechanisms as plausible.

What does it mean that homogeneous processes dominate the heterogeneous? Do you refer to the relative radiative forcing difference, the ice water content, ice crystal number concentration, frequency of occurrence of nucleation events?

Moreover, I am not sure whether figures from the latest ECHAM-HAM studies (e.g. Gasparini and Lohmann 2016, Gasparini et al., 2018) confirm your homogeneous vs. heterogeneous nucleation arguments. Homogeneous freezing seems to dominate only near the tropopause and over mountains. You could also cite Barahona et al., 2017, which shows somewhat consistent results with Gasparini and Lohmann 2016 in terms of homogeneous vs. heterogeneous freezing importance.

page 2, line 33/34 (and on page 33):

I don't think Sanderson et al., 2008 is looking at radiative balance of upper tropospheric clouds, but rather at the sensitivity of climate feedback to tuning parameters.

Also, Mitchell et al., 2008 look at differences in simulated climate by changing the particle shape distributions, affecting the fall velocities, and finally the radiative effects of clouds.

page 3, line 5

Liquid (or more precisely aqueous) sulphuric acid droplets CANNOT act as ice nucleating particles for heterogeneous freezing.

The increase in IC number concentration in Cirisan et al., 2013 is related to the presence of large sulphuric acid particles, which makes homogeneous freezing more favourable. Sulphuric particles at r<0.1 μ m only hardly nucleate ice crystals homogeneously due to the strong Kelvin effect. Stratospheric perturbations shift this distribution closer to sulphuric aerosol radii between 0.6 and 0.8, which were shown to be most susceptible for homogeneous freezing (see paragraph 2.3 of Cirisan et al.).

page 4, lines 5-10

The discussion seems to clearly highlight the thinning of cirrus in presence of a volcanic forcing. I think that by our current best knowledge we cannot give a conclusive answer on the influence of volcanic eruptions on cirrus clouds

frequency, microphysics, or radiative properties (e.g. Meyer et al., 2015 has a different conclusion from the study you cited).

page 7, line 18

Homogeneous freezing threshold is not constant, but should have some temperature (or, more precisely, water activity) dependence. Many parameterizations follow the Koop et al., 2000 results/formula. You have to therefore mention that important shortcoming, which might lead in most places to some overestimation of your homogeneous freezing probability, and the opposite at temperatures close to the homogeneous freezing temperature of water.

page 7, line 32-33:

Again, I don't think there is much evidence for the dominant role of homogeneous freezing. At most, you can mention that the relative importance of homogeneous vs. heterogeneous freezing is currently still very uncertain. The cited study with the message: "beware of the coating of dust, which decreases the ability to nucleate ice of several ice nucleating particles" (i.e. Cziczo et al., 2009) is not a proof of your statement!

page 10, line 8-10:

That's surprising; I would expect that the IWC at the lowest levels is dominated by detrained sources. Indeed, you might be just looking at IWC_{het}/IWC_{hom}, which is OK, but you need to mention in this case the missing and probably large convective IWC source in the tropics below about 12 km.

page 11, line 9-15:

I do not see much value in the comparison of your globally averaged ice crystal number concentration with a randomly picked study from a field campaign (which is, moreover, likely affected by the pre early 2000s retrieval problems due to ice crystal shattering, see Cziczo et al., 2014).

Again, I also do not see any reason to trust the MERRA+MODIS derived IC number concentrations on Figure 4.

Figure 12 and related text:

Again, the derived extinction from MERRA+MODIS does not add much of scientific value.

Same for Figure 13 b.

page 30, line 3-5:

Background clouds have a positive cloud radiative effect. That means they reflect less (if we assume all comes from SW), and not more!

If the solar radiation reaching top of the clouds decreases by, say, 1%, the amount of reflected SW radiation has to also decrease by the same relative value to first order (1% in this example).

Figure 14:

Why is the background cloud effect plotted only once? I guess it does change between the two cases.

Figure 15:

You never show that there is reduced water vapour transport to the upper troposphere? Prove it or remove it!

Also, I would like to see some evidence for the "convectively driven tropospheric cooling" before putting that in your summary sketch!

In summary, your schematic is a bit too complicated to be easily digested by an average reader. I think you can drop a few of the points, unless you prove them to be crucial in delivering your message.

page 31, lines 27-28:

Or maybe simply the water cycle slows down due to decrease of surface temperature, following Clausius-Clapeyron?

page 33, lines 14-16:

This is not really a good explanation of the SW adjustment. It is rather confusing to the reader. I thought you do not include cirrus in the "background clouds" effects based on your Figure 14, which shows the background effect separately from the effect on ice clouds.

References

Ackerman et al., 2008: Cloud detection with MODIS. Part II: Validation Bosilovich et al., 2015: MERRA-2: Initial Evaluation of the Climate

Cziczo et al., 2014: Sampling the composition of cirrus residuals

Duncan and Eriksson, 2018: An update on global atmospheric ice estimates from satellite observations and reanalyses

Hong et al., 2012: Estimating effective particle size of tropical deep convective clouds with a look-up table method using satellite measurements of brightness temperature differences

Koop et al., 2010: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions

Meyer et al., 2015: Did the 2011 Nabro eruption affect the optical properties of ice clouds?

Yang et al., 2007: Differences Between Collection 4 and 5 MODIS Ice Cloud Optical/Microphysical Products and Their Impact on Radiative Forcing Simulations

Zhang et al., 2010: Effects of ice particle size vertical inhomogeneity on the passive remote sensing of ice clouds