

Response to reviewer # 1

Reviewer comments are in bold. Author responses are in blue.

This is an interesting and timely study of how geoengineering in the form of stratospheric aerosol injection (SAI) would impact ice clouds in the upper troposphere (UT). Few papers have been published on this poorly understood aspect of SAI, so in that sense the paper is a welcome contribution to the geoengineering discussion.

We would like to thank the reviewer for his insightful comments and suggestions. We will try to address all of them below.

Overall comment:

However, the paper is in its current form very unclear when it comes to the representation of central processes in the ULAQ model, poorly structured, and full of incorrect/poor grammar. It needs a serious overhaul in all these respects. I further question the validity of the results presented, in light of the coarse resolution of the model, as well as its overly simplistic treatment of UT ice nucleation. I challenge the authors to justify why their results can be trusted despite these shortcomings. Below I've listed some additional major concerns I had about the paper, and thereafter some minor comments (typos, questions for clarification, etc.). I would like to see all of these concerns addressed before I will consider the paper suitable for publication in ACP.

- (a) A better, clearer and more complete presentation of the main processes governing ice particle formation in the ULAQ-CCM has been made in the revised manuscript.
- (b) An improvement in the manuscript structure has been made, following also a specific recommendation from the reviewer (see below).
- (c) Following the suggestion of both reviewers, an English technical editing of the manuscript has been done.
- (d) The ULAQ-CCM version adopted in this study uses a T21 horizontal resolution, which may be (in general) defined as a rather coarse horizontal resolution. On the other hand, it has been demonstrated in many previous published works that this is a fully acceptable resolution for studies focusing on stratospheric dynamics and transport, as well as on strat-trop exchange (e.g., Pitari et al., 2016a; Pitari et al., 2016b; Visioni et al., 2018). It is obviously possible to use higher horizontal resolutions, but this is not a strict physical requirement. Many model inter-comparison campaigns prove this (see for example SPARC-CCMVal-2 or the ongoing SPARC-CCMI: Morgenstern et al., 2017; Morgenstern et al., 2018; see also other referenced papers in the manuscript, where the ULAQ model scores well compared to other models with higher horizontal resolution). This is even more true in the case of UT ice formation, which is largely driven by sub-grid vertical motions in global composition models; the latter may explicitly predict only the large-scale dynamics and need to parameterize mesoscale vertical motions, even with higher horizontal resolutions. On the other hand, we believe that the use of a high vertical resolution is necessary to properly catch the time-latitude-longitude varying altitude of the tropopause and then the upper limit for UT ice formation. A proper vertical resolution is indeed adopted in the ULAQ model (568 m in pressure altitude). In this way, the different aerosol behavior above and below the tropopause altitude is also well caught in the ULAQ-CCM.
- (e) Cirrus ice formation in the ULAQ-CCM results from both homogeneous and heterogeneous freezing mechanisms and their competition. However, in our first draft of the manuscript we had decided to turn off the heterogeneous freezing mechanism, in order to focus on the SG

aerosol induced perturbation to ice formation from homogeneous freezing only. We acknowledge the specific point raised by the reviewer. The reduced updraft will affect ice formation from homogeneous freezing in a different way if ice particles may also form via heterogeneous freezing. This latter process, in fact, requires normally lower supersaturation conditions, both on mineral dust and black carbon particles. For this reason, we have performed again all our simulations with both mechanisms turned on, allowing their non-linear interaction. Results are substantially affected, with a resulting smaller indirect RF due to upper tropospheric ice changes induced by sulfate geoengineering. Looking at the revised results of our numerical experiments, we are really indebted with both reviewers for raising this specific scientific point, helping us in a more correct assessment of the ice perturbation due to SG aerosol and its indirect radiative forcing. In the manuscript, we now give a compact description of how ice particles are formed on both channels (HET-HOM), highlighting the major source of uncertainty in the parameterization of the heterogeneous freezing. A more robust scientific knowledge is present for the homogeneous freezing mechanism. In this case the numerical code adopted in the ULAQ model in the one well documented in the literature by Kärcher and Lohmann (2002) and Lohmann and Kärcher (2002), plus other subsequent studies, among which the most relevant for our present purposes is Kuebbeler et al. (2012).

Major comments:

1) With respect to the paper structure, I found it strange that in Section 2 ("ULAQ-CCM and setup of numerical experiments") some model results in response to SG are presented (in Fig. 3-6), but not until sections 2.1 and 2.2. are descriptions of the model treatment of stratospheric aerosols and ice clouds described. I suggest moving the presentation and discussion of model results until AFTER you've described the model, and to only put content in the various sections that is consistent with the section titles.

In the revised version of the manuscript we have shown all the model results after the sections where we describe the model, as suggested.

2) There is no discussion of the effect of additional SO₄ available for homogeneous nucleation in the SG cases, as evident in Fig. 7a. Why is the effect of what appears to be a tripling of SO₄ particles that can nucleate ice seemingly negligible? Please explain?

The increase of SO₄ in the upper troposphere due to the sulfate injection has a negligible effect on the rates of homogeneous freezing, as shown in Cirisan et al. (2013) (mainly because the number density of background SO₄ aerosols in the UT is already much greater than the number density of ice crystals). Furthermore, liquid supercooled sulfuric acid aerosols are inefficient IN for heterogeneous freezing; solid aerosol particles, mainly mineral dust and black carbon, may act as IN with different ice active fraction depending on aging processes and environmental conditions (e.g. Hendricks et al., 2011). For these reasons, changes in the UT population of sulfate aerosols are not expected to play a direct role in the changes in ice particle formation processes. It is actually the thermo-dynamical perturbation induced by lower stratospheric SG sulfate aerosols that may significantly impact the rate of ice formation via homogeneous freezing. This is indeed the central point of our study. In the revised manuscript, we have addressed this issue in a more in-depth way.

Minor comments:

Abstract: "Goal of. . ." should be changed to "The goal of.."

Changed.

Abstract: Don't understand what "coupled to" means in this context.

In previous experiments looking at sulfate geoengineering changes on UT ice, surface temperatures were kept fixed and only the LS warming was considered. In our case, in the experiment G4 both the surface cooling and the LS warming contribute to the modifications of the atmosphere dynamics.

Page 2, line 4, End sentence after "documented."

Done.

Page 2, line 9: Add "optimal" before "magnitude and location".

Added.

Page 2, line 23: Add reference for the claim that homogeneous nucleation normally dominates cirrus formation. "Supersaturation ratio" should be "saturation ratio".

Most of the literature considering geoengineering experiments (in particular, cirrus seeding) point out that most of the freezing is due to homogeneous processes, and that when including heterogeneous freezing processes, the differences are small. See for instance Gasparini et al. (2015), Gasparini et al. (2017), Storelvmo et al. (2014). We have added in the text these references and discuss better the relative weight of the two freezing mechanisms. Beyond geoengineering experiments, see also Kärcher and Lohmann (2002) for the specific point of homogeneous freezing normally dominating over the heterogeneous freezing of cirrus ice formation. Uncertainties in the latter case are discussed in Hendricks et al. (2011).

Page 2, line 31: "anyway" is not suitable here. "However" could be an alternative.

Corrected.

Page 2, line 32: cloud optical properties are also important here.

Added.

Page 4, line 1: This statement is confusing: sulphuric acid droplets are not ice nuclei. Please clarify. Furthermore, this statement is not very interesting unless you explain WHY there was no effect on RF.

As specified above in the response to the second major point, sulphuric acid liquid supercooled droplets are very inefficient ice nuclei. For the sake of increasing clarity, we have modified our sentence in the following way: "An upper tropospheric increase of sulfate aerosol number concentration is expected in SG conditions, due to gravitational sedimentation and large-scale transport of the particles below the tropopause from the LS. However, sulphuric acid liquid supercooled droplets are very inefficient ice nuclei (IN) for heterogeneous freezing. At the same time, the background number concentration of UT aerosols acting as nuclei for homogeneous freezing is already much higher with respect to the ice particle number density. For this reason, a negligible increase of the active IN population would be found in the UT, and the same would hold

true for the positive RF associated to a possible increase of ice particles from this effect, as Cirisan et al. (2013) conclude in their study.”

Page 4: Vertical velocity is important for cirrus formation not primarily because it transport water vapour to the UT, but because it controls the adiabatic cooling rate and thus supersaturation, for a given water vapour content.

We have changed the sentence accordingly.

Page 4, line 14-17: Catastrophic grammar.

Grammar adjusted.

Figure 1: This figure is confusing and not well explained. I don't find it particularly helpful at this stage of the paper, but it could be good as a final figure summarizing the findings in the paper.

This figure has been moved to the final part of the manuscript, as well as Figure 2.

Page 5, line 5: Ash dust is not the same.

Corrected.

Page 6, line 1: Sassen et al. (2008) is a paper on cirrus coverage seen by CALIPSO, so I don't see how that could possibly address UT ice changes.

Yes, it was a mistake. The reference we were intending to put was Sassen et al. (1995), where possible changes in cirrus are discussed in relation to the Pinatubo eruption. We have now corrected this in the revised manuscript.

Page 6/Table 1: The horizontal resolution is extremely coarse - how can we have confidence in changes driven by dynamics in this context?

As explained above in the response to the overall comment of the reviewer, the ULAQ-CCM version adopted in this study uses a T21 horizontal resolution, which may be (in general) defined as a rather coarse horizontal resolution. On the other hand, it has been demonstrated in many previous published works that this is a fully acceptable resolution for studies focusing on stratospheric dynamics and transport, as well as on strat-trop exchange. Many model inter-comparison campaigns prove this (see for example SPARC-CCMVal-2 or the ongoing SPARC-CCMI). In addition, UT ice formation is largely driven by sub-grid vertical transport processes in global composition models; the latter may explicitly predict only the large-scale dynamics and need to parameterize mesoscale vertical motions, even with higher horizontal resolutions.

Page 7, line 4: Clumsy and confusing statement. Suggest writing: ...a negative anomaly in the Arctic region that is approximately 1 K larger than that of high southern latitudes.

We have rephrased it as the reviewer suggested.

Page 7, line 7: What do you mean by “increasing atmospheric stabilisation”? Do you mean “increasing atmospheric stability?”

Yes, we have corrected this.

Page 7, line 16-18: The Antarctic warming is not statistically significant, so I don't see the point in discussing it.

Although the reviewer remark is correct, we would like to keep this short discussion as a justification for the large variability of SST changes at high latitudes. A slight modification has been made by writing: "Although not statistically significant, the SG induced warming..."

Page 8, line 2: Is the vertical velocity change mainly caused by changes to TKE, or also due to large-scale (resolved) velocity changes. If TKE is very important here, I would like to see vertical profiles of TKE for both simulations.

In the revised manuscript, we have explicitly included the Lohmann and Kärcher (2002) formulation for the vertical velocity (Eq. 5). In a first approximation, w in the UT is close to $TKE^{0.5}$ and the vertical velocity perturbation is dominated by changes of this latter term, so that we believe that inclusion of a new figure in the revised manuscript does not add much. We attach here the vertical profiles of w in G4 and Base experiments (Fig. R1_1), as well as the vertical profile of w changes [G4-Base], comparing the results with and without the large-scale contribution to w . It is clear that TKE changes greatly control the SG perturbation of UT updraft. The TKE vertical profile asked by the reviewer is implicit in panel (b) of the Fig. R1_1, due the w_{TKE} formulation.

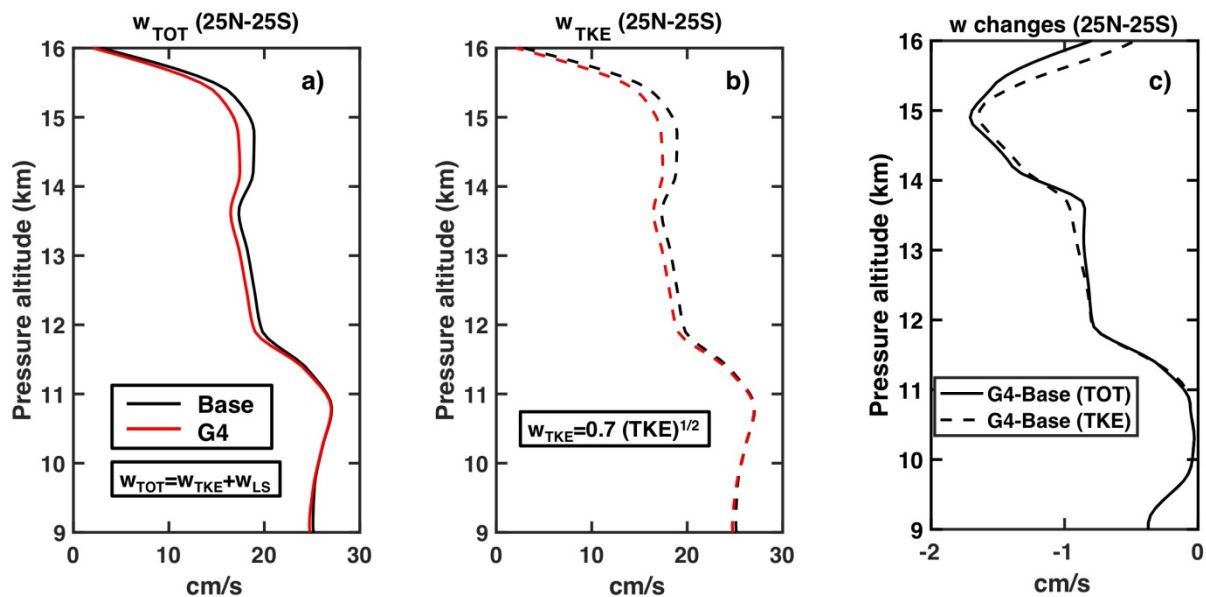


Fig. R1_1. Average upper tropospheric tropical profiles of the vertical velocity w (cm/s) in G4 and Base experiments (years 2030-39). (a) Total vertical velocity calculated as $w_{TOT} = w_{TKE} + w_{LS}$ (where w_{TKE} indicates the mesoscale component calculated as a function of TKE and w_{LS} indicates the large-scale model-resolved component). (b) Vertical velocity component w_{TKE} alone, calculated as $w_{TKE} = 0.7 \times (TKE)^{1/2}$ (see Lohmann and Kärcher, 2002; see also Eq. 5 in the revised manuscript). As expected, w_{TKE} dominates in w_{TOT} (c) Vertical velocity changes G4-Base for w_{TOT} (solid line) and w_{TKE} only (dashed line) (of the order of 5% in the tropical UT). Very little changes are produced in the large-scale component under SG conditions.

Page 9, lines 19-20: Discuss here the uncertainty associated with cloud ice in MERRA, which uses highly uncertain cloud parameterisations and incorporates very few ice cloud

observations in its reanalysis. It would be better to use CALIPSO/CloudSat retrievals of ice cloud properties.

In the revised manuscript, we have added some discussion on the uncertainties of the datasets we have used for comparison with our results. We have decided to use a reanalyses dataset such as MERRA-2 also considering the large uncertainties in the satellite retrieval datasets (see for instance Zhang et al., 2010; Duncan and Eriksson, 2018) and their availability.

Page 10, line 3: Given how central UT vertical velocities are to this paper, you need to be clearer about how the calculation of vertical velocity is done, i.e. include equation for vertical velocity as a function of TKE, and clearly state if you put any upper/lower bounds in it.

In the revised manuscript, we have explicitly included the Lohmann and Kärcher (2002) formulation for the vertical velocity (Eq. 5) (see also Fig. R1_1 above). No imposed bounds to w are considered and its time-spatial variability is clearly shown in Figure 6 of the original manuscript.

Page 10, line 4: What justifies the assumption that cirrus clouds form only via homogeneous nucleation? That seems to be in stark contrast to papers that report that cirrus clouds appear to form mainly through heterogeneous nucleation (e.g., Cziczo et al., 2013).

Please refer to our reply above, regarding the homogeneous - heterogeneous freezing mechanisms, in response to the reviewer overall comment. In the revised manuscript, we discuss the major source of uncertainties and add a caveat regarding the presence of different opinions available in the literature, like the ones the reviewer suggested.

As specified above, we have taken the reviewer criticism under serious consideration and decided to redo our numerical simulations with both freezing mechanisms turned on, allowing their non-linear interaction. Results are substantially affected, with a resulting smaller indirect RF due to upper tropospheric ice changes induced by sulfate geoengineering. Looking at the revised results of our numerical experiments, we are really indebted with both reviewers for raising this specific scientific point, helping us in a more correct assessment of the ice perturbation due to SG aerosol and its indirect radiative forcing.

Page 13, line 15: Remove “from”.

Removed.

Page 16, line 13-14: This is inaccurate - homogeneous nucleation sets in at approximately 238K, but NOT through "water vapour freezing", but rather through the spontaneous freezing of small solution droplets.

Corrected accordingly.

Page 16, line 18-19: This is an outdated view (and references that back this claim are not provided) - the current understanding is that a majority of cirrus clouds form via heterogeneous nucleation.

Please see above. Heterogeneous nucleation would dominate only if the locally available IN (mostly BC and mineral dust) would have a high ice active fraction ($>\sim 10\%$). These values, however, although being measured in laboratory studies for mineral dust close to the homogeneous freezing threshold (Field et al., 2006; Möhler et al., 2006; Welti et al., 2009), are most probably

highly overestimated in the real atmosphere, due to rapid aging of dust particles (as well as BC) through sulfate coating (Hendricks et al., 2011). We acknowledge (and added in the revised manuscript) the counterpoint that studies such as Cziczo et al. (2013) show that the heterogeneous freezing may dominate over the homogeneous, in the formation of UT ice particles. However, we believe there is plenty of literature showing through both modeling and in-chamber experiments the huge uncertainties relative to our understanding of UT ice formation through heterogeneous freezing, in particular regarding the available aerosol population that is actually able to form ice in the upper troposphere (ice active fraction).

For instance, regarding black carbon, laboratory measurements demonstrate that the ice active fraction (f) ranges between 0.1% and 1% (Koehler et al., 2009), which means that only a very small fraction of the available black carbon particles in the UT can act as IN. Considering the rapid BC aging in the real atmosphere (due to sulfate coating), $f \sim 0.1\%$ may be probably considered as an upper limit for the ice active fraction, although a clear picture has not yet emerged for the factors which actually control f for a given type of atmospheric IN (Hendricks et al., 2011), thus producing a significant level of uncertainty in the present knowledge of UT ice formation via heterogeneous freezing. For mineral dust the uncertainty is even higher, with f ranging between 0.1% and 10%, although it might be even lower (Minikin et al., 2003; Cziczo et al., 2009).

Those measurements are the only one that, as modelers, we can take into account when considering which fraction is to be used in our simulations (in our experiments we chose $f=0.25\%$ for BC and 1% for mineral dust, following the best recommendations of Hendricks et al., 2011, see Eq. 1 in the revised manuscript).

Fig. 8: Again, I do not think of MERRA as the most appropriate data set for validation of the simulated UT ice.

See response above.

Page 16, line 21 (and throughout the manuscript): The standard terminology is “ice mass mixing ratio”, not “ice mass fraction” which can be misleading.

Following the suggestion, we have changed it everywhere in the manuscript.

Page 16, line 8 - 15: The description of how UT ice clouds form is extremely unclear. How is cloud cover determined? What probability distribution for supersaturation is used, and how does it relate to TKE. A lot of essential information is left out here.

We disagree on this point. We have clearly stated our simplified probabilistic approach adopted for supersaturation, with a normal (Gaussian) distribution for the UT relative humidity: “For the ice supersaturation ratio, we adopt a simplified probabilistic approach, starting from the knowledge of climatological frequencies of the UT relative humidity (RH_{ICE}), from which a mean value and a standard deviation can be calculated, assuming a normal distribution”. We are aware that this represents an important model simplification, and in fact we started our discussion with the above clear statement.

Page 18, line 4: “each thick” is not correct English.

Corrected.

Page 18, line 8: What do you mean by “we are only considering sub-visible clouds”?

The sentence has been modified as follows: “This should not surprise, in principle, due to the fact that vertical velocities calculated as a function of TKE do not normally exceed 30 cm/s, so that events leading to thick cirrus formation are not considered.”

Fig. 10: Is the ice crystal number density calculated only when there is a cloud (i.e. an in-cloud average), or is this an average over both cloudy and cloud-free grid-boxes? The former quantity is certainly of most interest and more directly comparable to field measurements.

We always refer to averages weighted with the probability to have cirrus formation ($\sim P_{\text{HOM}}$). The reviewer is right in saying that an in-cloud average would be more directly comparable to field measurements. That’s why we used our P_{HOM} to make this type of comparison in a meaningful way. In the original manuscript, we wrote: “Using these P_{HOM} values, it is possible to scale a n_i value measured in the mid-latitude airborne campaign of Ström et al. (1997) during a young cirrus formation, in order to derive an average climatological value to be considered consistent with our modeling approach. They measured a mid-latitude ice concentration value $n=0.3 \text{ cm}^{-3}$ in a young cirrus cloud at $T=220 \text{ K}$ and $p=320 \text{ hPa}$. If we scale this result with our corresponding $P_{\text{HOM}}=12\pm3\%$, a “climatological-mean” value $n=0.025\pm0.005 \text{ cm}^{-3}$ is obtained, close to our model predicted value of $0.031\pm 0.008 \text{ cm}^{-3}$.”

Page 18, line 10-11: Neglecting heterogeneous ice nucleation would lead to an overestimate of ice crystal number, because you are not able to represent the competition between heterogeneous and homogeneous nucleation that will in some cases lead to a suppression of homogeneous nucleation and therefore a reduction in ice crystal number density. In other words, that cannot explain the disagreement with MERRA+MODIS seen in Fig. 9.

The reviewer is perfectly right. As explained above in detail, our new results confirm this. Again we thank both reviewers for raising this point and providing us a strong scientific argument to redo our numerical experiments. The following sentence has been deleted: “In addition, ice formation from heterogeneous freezing on active IN, as mineral dust particles for example, is not taken into account in our modelling approach.”

Page 26, line 7-9: How can you be confident about the radiative effect when the model consistently produces ice clouds that are optically too thin? This could bias especially the LW cooling effect of cloud thinning.

Our confidence comes from comparing our results to previous findings (as in Kuebbeler et al., 2012, Gasparini et al., 2017). In addition, the reviewer point is rather unclear, in the sense that the ice OD for G4, G4K and Base simulations is not small in absolute values. We may then calculate the ice radiative effects both on SW and LW, once appropriate Mie scattering parameters have been derived, using a correct wavelength-dependent refractive index (Warren 1984; Warren and Brandt, 2008; Curtis et al., 2005) and the calculated particle size distribution. Results of our radiative transfer code have been successfully compared with those of Schumann et al. (2012) under similar conditions. We have added in the paper the appropriate references.

General comment: Friberg et al. (2015) seem to qualitatively support your findings based on analysis of cirrus cloud reflectance changes after volcanic eruptions, so that would be a good paper to cite.

We have read the paper suggested by the reviewer and had the occasion to speak with the lead author. In the revised manuscript, we now briefly discuss their conclusions and have added the appropriate reference.

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