Reviewer 1

We thank the reviewer for the time spent evaluating our study and for the valuable comments and suggestions, which helped us to substantially improve the manuscript. To address the reviewer's comments, some supplemental experiments were carried out. These supplemental experiments were run for 3 model years, and the last 2 years were used in the analyses. We hope that the revised manuscript and our response to the comments are satisfactory. The reviewer's comments are in italics, and our responses are in standard font below. In the last part of this response, we attached the revised manuscript with the main changes marked in blue.

General Assessment:

The reviewed manuscript presents climatic impacts of various seeding strategies in CAM5 general circulation model. In particular, the authors focus on a newly developed optimal seeding method that injects just the right amount of ice nucleating particles (or ice crystals, as suggested in the text) to prevent the formation of homogeneous freezing. Interestingly, seeding was found to modify liquid and mixedphase clouds in ways that counteract part of the climatic cooling effect at cirrus levels. Such adverse effects can be limited by seeding only areas with solar zenith angles larger than 12°.

The manuscript is nicely structured, has a clear message, and presents the model results in a convincing way. The optimal seeding method is an innovative new way of implementing cirrus cloud seeding in ice nucleation schemes. I recommend publication after minor revision.

Reply: We do appreciate the positive comment.

General comments:

1.) Why is homogeneous freezing switched off at temperatures colder than -68°C? I don't think there is enough evidence from observations for such a drastic modelling choice. Does this decrease the uppermost tropospheric ice crystal number and in such way brings the model closer to the observations? Does this condition influence upper tropospheric RHice?

Would cirrus clouds have a stronger warming effect if you allowed freezing also under the coldest temperatures? Would that increase the radiative impact of cirrus seeding?

Reply: At temperatures below 205 K, the observed ice number concentration (N_i) is usually in the range of 10-80 L^{-1} (Krämer et al., 2009), whereas the modeled N_i without switching off homogeneous freezing is usually in the range of 50-2000 L^{-1} (Shi et al., 2015). Because the theory for homogeneous freezing of aqueous aerosols predicts relatively high N_i for cold environments, the N_i from homogeneous freezing should be limited (Jensen et al., 2010). Some recent studies have shown that aerosols rich in organic matter may become glassy (i.e., potential ice nuclei particles) below the glassy-transition temperature (~205 K), and then prevent homogeneous nucleation (Murray et al., 2010; Shiraiwa et al., 2017). Furthermore, a previous study showed that the modeled N_i would be close to the in-situ observation if homogeneous freezing at temperatures colder than the glassy-transition temperature were artificially switched off (Fig. 6 of Shi et al., 2013). In this study, the main purpose of this artificial setting is to make the modeled N_i to be close to observations at temperatures below 205 K. We mentioned this purpose in the revised manuscript.

If homogeneous nucleation is allowed under 205 K, N_i increases over there. Correspondingly, cirrus clouds have a stronger warming effect. However, the cooling effect caused by cirrus seeding is decreased (i.e., weaker cooling effect). Figure S1 shows the comparison between the experiments with (REF and OPT experiments) and without the limitation (REFnolimit and OPTnolimit experiments) of homogeneous freezing under 205 K (hereafter limitation). As expected, the ice cloud radiative effect (iCRE) from the REFnolimit experiment (6.68 W m^{-2}) is slightly increased (i.e., stronger warming effect) compared to that from the REF experiment (6.48 W m^{-2}) because N_i is increased under 205 K (not shown). The cooling effect from ice clouds (ΔiCRE) caused by cirrus seeding without the limitation (OPTnolimit − REFnolimit, −3.16 W m^{-2}) is also slightly stronger than that with the limitation (OPT − REF, −3.04 W m^{-2}). The cirrus seeding effectiveness from the OPTnolimit experiment (49.91%) is close to that from the OPT experiment (49.33%). However, the global cooling effect (ΔCRE, from all clouds) from the OPTnolimit experiment (OPTnolimit − REFnolimit, −1.11 W m^{-2}) is obviously weaker than that from the OPT experiment (OPT − REF, −1.35 W m^{-2}). As introduced in the manuscript, cloud seeding leads to a significant warming effect of liquid and mixed-phase clouds, which counteracts the cooling effect of cirrus clouds. This counteraction becomes stronger in the OPTnolimit experiment.

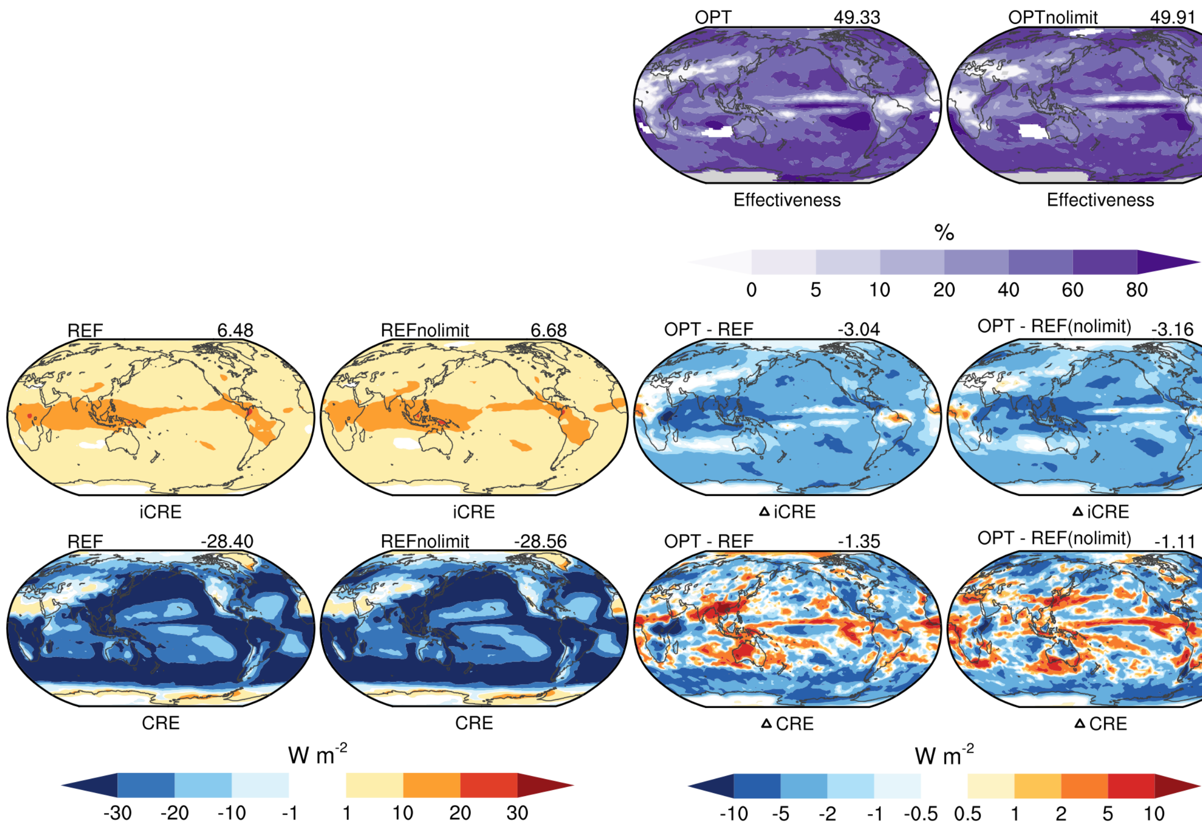


Figure S1: The annual mean spatial distribution of the ice cloud radiative effect (iCRE, second row) and all cloud radiative effect (CRE, third row) from the REF and REFnolimit experiments (left panel) and the differences from the OPT and OPTnolimit experiments (right panel) with respect to the REF and REFnolimit experiments. The cirrus seeding effectiveness are shown in the first row. Global mean values are shown in the upper right corner. Notably, only 2-year simulation results from each experiment are analyzed here.

2.) What is the spatial and temporal variability of seeded ice crystals in the OPT scenario within a certain region/time? E.g. does the optimal seeded ice crystal number remain constant on the 0.5/1/2/7/24/90-day timescale within a certain area? Would the OPT seeding strategy be feasible in real world or in parts of the world, or should we think of it as a purely academic experiment?

Also, please specify whether you seed at every model timestep in INP20 and INP200 strategies. Would the OPT scenario with a decreased seeding frequency still deliver a significant cooling effect?

Reply: Thank you for the comments. Figure S2 shows the number concentration of seeding ice crystals

($N_{seedopt}$) on the 0.5-, 2-, and 90-day timescales over three different model grids. The $N_{seedopt}$ over different regions/times are different. Notably, $N_{seedopt}$ is a function of the ambient atmospheric condition and the given radius of seeding ice crystals (R_{seed}). Therefore, $N_{seedopt}$ can be calculated from the current atmospheric conditions or roughly calculated based on the numerical weather forecast for the next few hours. How to determine $N_{seedopt}$ may not be the main barrier to the feasibility of the OPT seeding strategy in the real world. However, many aircrafts are needed to add ice crystals at specified times and locations, which is the main barrier to the OPT seeding strategy. This study is a pure academic experiment. The main purpose of this study is to estimate the potential cooling effect of cirrus thinning.

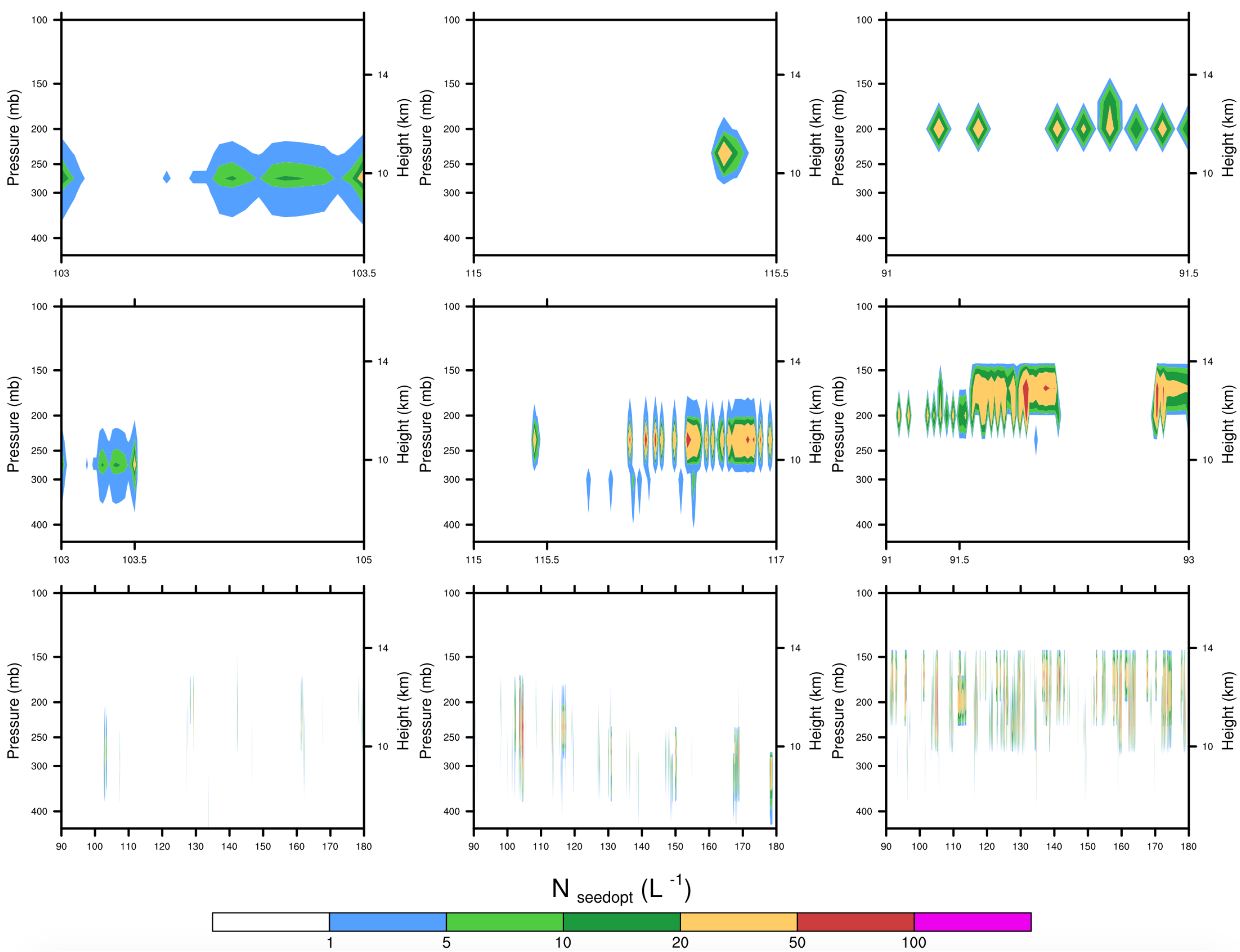


Figure S2: The vertical distribution of $N_{seedopt}$ over three model grids, 120 °E and 45 °N (leftmost column), 160 °E and 30 °S (middle column), and 180 °E and 4 °N (rightmost column) on the 0.5- (upper), 2- (middle), and 90-day (lower) timescales. The horizontal coordinate axis is the model time (unit: day).

In the INP20 and INP200 experiments, ice nuclei particles (INPs) are seeded at every model time step, but the seeding INPs only impact the ice nucleation process. This seeding strategy was emphasized in the experimental setups in the revised manuscript.

Similar to the OPT experiment, one more experiment (Reduce experiment) that randomly canceled the seeding operation was carried out. The seeding frequency from the Reduce experiment is approximately half of that from the OPT experiment (Fig. S3). In the Reduce experiment, ΔiCRE and ΔCRE are −1.59 and −0.91 W m^{-2}, respectively (Fig. S4). The global cooling effects from the Reduce experiment are weaker than those from the OPT experiment (the 2-year averaged ΔiCRE and ΔCRE are −3.04 and −1.35 W m^{-2}, respectively, Fig. S4).

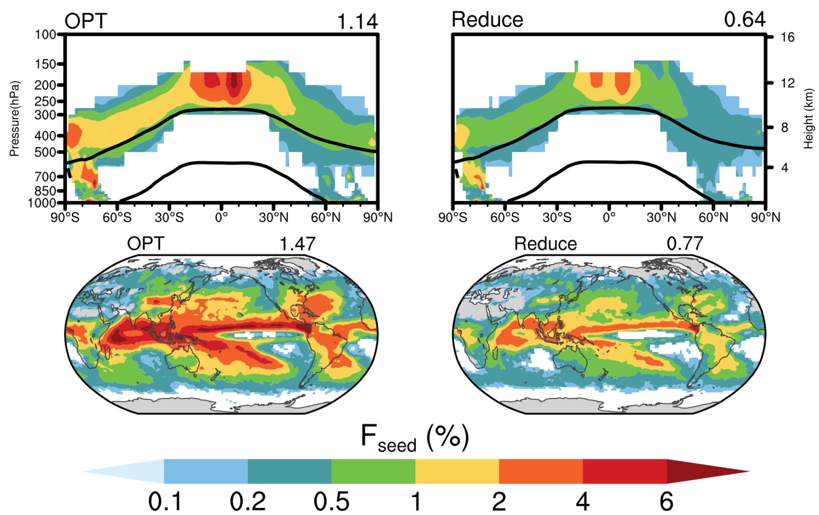


Figure S3: Annual zonal mean and 233 hPa spatial distribution of seeding frequency (F_{seed}). The names of the experiments are shown in the upper left corner, and global mean values are shown in the upper right corner. Notably, only 2-year simulation results from each experiment are analyzed here.

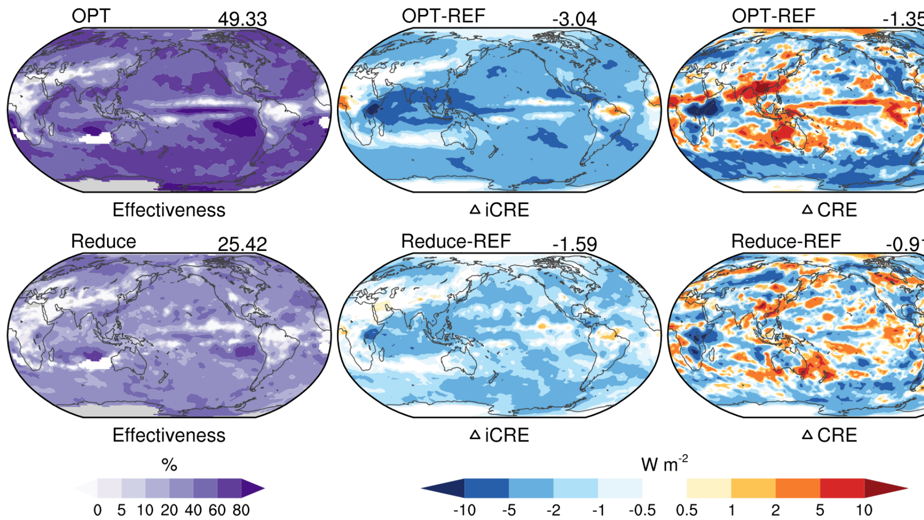


Figure S4: The annual mean spatial distribution of differences in iCRE and CRE from the OPT and Reduce experiments with respect to the REF experiment. The cirrus seeding effectiveness are shown in the leftmost column. Global mean values are shown in the upper right corner. Notably, only 2-year simulation results from each experiment are analyzed here.

Specific comments:

3.) page 1, line 20: conserving energy => probably meant "saving energy"?

Reply: Thanks for the comment. Done.

4.) page 1, line 23: …as a back-up tool ~~to~~ against…

Reply: Thanks. Done.

5.) page 2, line 30: The sentence "…which allows more longwave radiation to escape into space so that cool the Earth…" should be rewritten.

Maybe: which allows more longwave radiation to escape to space, leading to a cooling effect on the planet.

Reply: Thanks. In the revised manuscript, the sentence was rewritten based on this comment.

6.) page 2, line 39: "…and even the strongest cooling effect may not be ideal" What do you mean with this sentence, please explain!

Reply: Thank you for this comment. Previous studies have shown that the global cooling effects with fixed seeding number concentrations are not strong enough even for their strongest cooling effects (usually above −1.0 W m^{-2}). In the revised manuscript, the sentence was rewritten.

7.) page 2, lines 60-62: The sentence on lines 60-62 sounds a bit weird and may need to be rewritten. A suggestion: Therefore, seeding the clouds with a few INPs can prevent the Sice to reach the threshold needed for….

Reply: Thanks. In the revised manuscript, the sentence was changed as suggested: "Therefore, seeding the clouds with a few INPs (usually less than 100 L^{-1}) can prevent S_i from reaching S_{ihom}".

8.) page 3, line 63-64: Not sure if we can draw a direct connection between decreased ice crystal number concentrations and the name "cirrus thinning". I believe it would be more intuitive for the reader to add the intermediate step of decreasing the cirrus cloud optical depth (as a result of less Ni), which is probably where the name thinning is coming from.

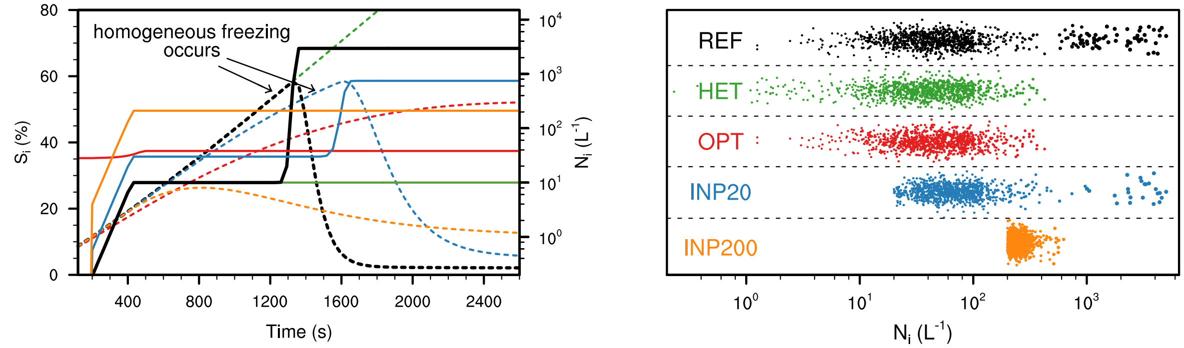
Reply: Thank you for this helpful comment. To improve the readability, the sentence was rewritten in the revised manuscript: "As a result, the in-cloud IC number concentrations (N_i) are usually decreased, which decrease the cirrus cloud optical depth (i.e., cirrus thinning)".

9.) page 3, line 93: Could you add a reference for the sentence starting with "Notably, in terms…"

Reply: Thanks. Some references (Pruppacher and Klett, 1998; Kärcher et al., 2006; Shi et al., 2015) were added in the revised manuscript.

10.) Fig.1: Why does the dashed line representing Si for HET simulation stop at about 60%? Shouldn't it continue increasing even after that point?

Reply: Yes, the S_i for HET simulation continues to increase throughout the process. In the revised manuscript, the dashed line representing S_i for the HET simulation stops at 80% (i.e., the upper limit of S_i in the figure). The updated figure is as follows:



11.) page 5, lines 136-137: I am confused by the two sentences here. The first one ("In the INP20 and INP200…") suggests you implemented seeding by simply increasing the N dust by 20/200 particles L-1. The sentence "Note that Ndust…" suggests exactly the opposite. So that you keep the N dust unchanged and you modify the Nseed. Could you therefore explain more thoroughly how did you introduce the seeding particles?

Reply: Thank you for this comment. We apologize for the confusion. Note that, "N_{dust} used for driving ice nucleation parameterization" and "N_{dust} in the aerosol module" could be two different variables. In the default model, the "N_{dust} used for driving ice nucleation parameterization" is same as the "N_{dust} in the aerosol module". In the INP20/200 experiments of this study, the "N_{dust} in the aerosol module" is not changed, but the "N_{dust} used for driving ice nucleation parameterization" = the "N_{dust} in the aerosol module" + 20/200 particles L^{-1}. To avoid confusion, the two sentences were rewritten in the revised manuscript.

12.) page 8, lines 97-98: Does this mean that in CAM model a substantial fraction of mixed-phase clouds is formed by sedimenting cirrus?

Reply: Thank you for this comment. Under the condition of 300 hPa and −35 °C, the sedimentation velocity of the 10 μm ice crystal is ~150 m $hour^{-1}$ (~ 4 km day^{-1}), and the sedimentation velocity of the 50 μm ice crystal is ~700 m $hour^{-1}$ (~ 18 km day^{-1}). The sedimentation process certainly influences the evolution of cirrus clouds. As shown in Fig. S5, the ice mass mixing ratio tendency caused by sedimentation (QISEDTEN) is negative at cirrus layers but positive at mixed-phase layers. Note that, ice crystals would sublimate if they fell into clear-sky areas. Therefore, Fig. S5 cannot indicate that a substantial fraction of mixed-phase clouds is formed by sedimenting cirrus. To avoid the misleading implication that sedimentation is the main mechanism for mixed-phase clouds formation, analyses of the possible reason for the increase of IWC at mixed-phase layers were added in the revised manuscript as follows: "In the HET and OPT experiments, both N_i and IWC in middle and lower mixed-phase clouds are obviously increased. The main reason might be that the deep convective activity becomes more vigorous because cirrus thinning reduces atmospheric stability via the radiative budget. The ratio of ice to total cloud condensate detrained from the convective parameterizations is a linear function of temperature between −40 °C and −10 °C (Morrison and Gettelman, 2008). Furthermore, the ICs can grow through the Bergeron process in mixed-phase clouds (Morrison and Gettelman, 2008). This might be the reason that the relative increases in IWC in mixed-phase clouds are stronger than the relative increases in N_i".

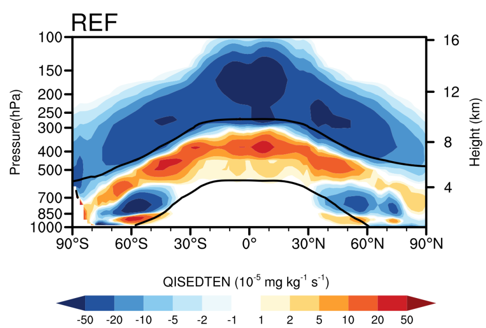


Figure S5: Annual zonal mean distribution of the ice mass mixing ratio tendency caused by sedimentation (QISEDTEN) from the REF experiment.

13.) page 8, line 200-201: Why - Are the regions where IWP increases connected to a more vigorous deep convective activity?

Reply: Yes, the regions where IWP increases are connected to more vigorous deep convective activity. Notably, the cloud condensate detrained from the convective parameterizations is considered as ice below −40 °C. More importantly, over these regions, the decreases of IWP induced by cirrus seeding are weak.

14.) page 10, line 239: "…is used to show how many proportions of iCRE are eliminated…" please rewrite, for instance as: …is used to show what proportion of iCRE is eliminated…

Reply: Thank you for this comment. In the revised manuscript, the sentence was changed as suggested.

15.) page 12, title 3.2: It's probably better to use "with" instead of "regarding".

Reply: Thanks. Done. Furthermore, we checked this expression throughout the manuscript.

16.) page 12: Another motivation for R10 experiment is probably the smaller mass of the 10 micron ice crystals, compared with the 50 micron one (?)

Reply: The total mass of seeding ice crystals from the R10 experiment (seeding ice crystal is 10 μm) is significantly smaller than that from the OPT experiment (seeding ice crystal is 50 μm), even if the number concentration of seeding ice crystals from the R10 experiment is twice as high as that from the OPT experiment. In this study, it is assumed that the seeding ice crystals can be made from ambient atmospheric water vapor. Thus, the transportation problem is not considered.

17.) page 15, line 350: regarding the => with the/using the

Reply: Thanks. Done.

18.) page 15, line 353: the global cool effect => the global cooling effect

Reply: Thanks. Done. We also carefully checked the entire manuscript for grammatical and formatting errors.

19.) page 15, line 354-355: The sentence starting with "Because there are…" should be rewritten. Maybe: Avoiding seeding over low-latitude regions can limit some warming effects due to changes in mixed-phase and liquid clouds and thus lead to a more pronounced global cooling effect.

Reply: Thanks. In the revised manuscript, the sentence was changed as suggested.

20.) page 16, line 360: maybe better: The overall cooling effect was maximized when using a solar zenith angle threshold of 12°.

Reply: Thanks. In the revised manuscript, the sentence was changed as suggested.

21.) page 16, line 363: the sentence "It is still possible…" should be reworded. Maybe: The global cooling effect can thus be maximized when limiting seeding to most suitable regions and times of the year.

Reply: Thanks. In the revised manuscript, the sentence was reworded as suggested.

22.) page 16, line 364: "…in the next step" => in our future work.

Reply: Thanks. Done.

23.) page 17, line 395: efficient => effective (?)

Reply: Thanks. Done.

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Shiraiwa, M., Li, Y., Tsimpidi, A., Karydis V., Berkemeier T., Pandis S., Lelieveld J., Koop T., and Poeschl U.: Global distribution of particle phase state in atmospheric secondary organic aerosols. Nat Commun., 8, 15002, https://doi.org/10.1038/ncomms15002, 2017.

Reviewer 2

We appreciate the thorough review and constructive comments, which have helped us to substantially improve the quality of the manuscript. We hope that the modified manuscript and our responses to the comments are satisfactory. The reviewer's comments are in italics, and our responses are in standard font below. In the last part of this response, we attached the revised manuscript with the main changes marked in blue.

1.) This manuscript documents experiments with variable ice nucleation number, by adjusting the ice concentration in a GCM to prevent homogenous freezing and thin cirrus clouds as a form of geo engineering. The manuscript is well written and thought out. It should be publishable in ACP with minor revisions.

My major concern is a lot of the effects are from liquid and mixed phase clouds that the method is not directly perturbing. Or should not be directly perturbing. But the mechanism is not explained well in the text. It is not clear if the compensating effects for mixed and liquid clouds from adding ice crystals to pure ice clouds is an artifact of the model, or has a solid physical basis. This should be detailed further in the discussion of the cases and the conclusions. It may require a bit more in depth analysis.

Reply: Thank you very much for your kind statement. We totally understand the reviewer's concern about the compensating effect (i.e., warming effect of liquid and mixed-phase clouds) caused by cirrus seeding. Gasparini et al. (2017) pointed out the compensating effects induced by cirrus thinning. There seems to be a relatively solid physical reason that cirrus thinning reduces atmospheric stability via the radiative budget and convective activity becomes more intense (Kristjánsson et al., 2015; Storelvmo and Herger, 2014), which would consume more cloud water and lead to the warming effect of liquid clouds (Gasparini et al., 2017; Rapp et al., 2011). The warming effect of liquid clouds in our study (0.94 ± 0.11 W m^{-2} from the OPT experiment) is similar to that reported by Gasparini et al. (2017; Table 5, 0.96 ± 0.25 W m^{-2} from the ECHAM-HAM model simulation with seeding 1 ice nuclei particle L^{-1} of 50 μm). However, the warming effect of mixed-phase clouds in our study (1.09 ± 0.11 W m^{-2}) is several times stronger than that from their results (Table 5, 0.15 ± 0.10 W m^{-2}). In our study (using the CAM model), the warming effect of mixed-phase clouds mainly comes from the increasing longwave component (not shown), which is consistent with the increased ice water content (IWC) in mixed-phase clouds. The main reason for the increased IWC might be that the convective detrainment is obviously increased due to cirrus thinning. The ratio of ice to total cloud condensate detrained from the convective parameterizations is a linear function of temperature between −40 °C and −10 °C (Morrison and Gettelman, 2008). Furthermore, ice crystals can grow through the Bergeron process in mixed-phase clouds (Morrison and Gettelman, 2008). The warming effect of mixed-phase clouds induced by cirrus thinning seems more sensitive to the cloud-related scheme used in the climate model. In the reversed manuscript, we added more analyses about the compensating effect (i.e., the warming effect of mixed-phase and liquid clouds) in Section 3. In the discussion section, we clearly pointed out that the climatic response to cirrus seeding is complex and might differ among different climate models and seeding methods. The compensating effects introduced in this study are derived from the atmosphere-only simulations with prescribed ocean surface conditions, the coupled model simulations might show different results (e.g., Gasparini et al., 2017).

Specific comments:

2.) Page 1, L11: is cooling positive or negative? Do the values in lines 10 and 11 with different signs represent the same direction? Please clarify: if the same direction then should be the same sign.

Reply: We thank the reviewer for pointing this out. Cooling should be negative even if it has been declared a cooling effect. A negative sign was added in the revised manuscript. Furthermore, we have checked the signs throughout the manuscript and emphasized this issue in the revised manuscript.

3.) Page 1, L14: cirrus clouds typically do not contain liquid, so why would cirrus seeding impact mixed phase clouds?

Reply: Thank you for this comment. Cirrus seeding can directly reduce the cirrus clouds and then indirectly impact mixed-phase and liquid clouds via different mechanisms. For instance, convective activity becomes more intense because cirrus thinning reduces atmospheric stability via the radiative budget, and the increased convective activity could impact liquid and mixed-phase clouds.

4.) Page 1, L16: could yield. Also, what sign is intended here?

Reply: Thanks. Done. The cooling effect is always quantified by a negative value in the revised manuscript.

5.) Page 3, L80: this seems like a pretty significant change to shut off homogeneous freezing below 205K. What is the impact of that?

Reply: Thank you for this comment. At temperatures below 205 K, the observed ice number concentration (N_i) is usually in the range of 10-80 L^{-1} (Krämer et al., 2009), whereas the modeled N_i without switching off homogeneous freezing is usually in the range of 50-2000 L^{-1} (Shi et al., 2015). In this study, the main purpose of this artificial setting is to make the modeled N_i to be close to observations at temperatures below 205 K (Shi et al., 2013). We mentioned this purpose in the revised manuscript.

6.) Page 4, L97: I think the formulas are an important part of the study, and should be in the main text.

Reply: Thanks for this suggestion. The flexible seeding method used for estimating the potential cooling effect of cirrus thinning includes the optimal seeding number concentration and the flexible seeding strategy. In the main text, we focus on introducing the advantages of the flexible seeding method, which are important for estimating the potential cooling effect of cirrus thinning. The formulas for calculating the optimal seeding number concentration provided by this study depend on the ice nucleation parameterization used in the climate model. However, the design idea of the optimal seeding number concentration is not model-dependent. For instance, some ice nucleation parameterizations provide the critical number concentration (N_{lim}) of ice nuclei particles (INPs) for the only heterogeneous freezing scenario (e.g., Barahona and Nenes, 2009). N_{lim} could also be used as the optimal seeding number concentration with INPs. Therefore, we prefer to focus on the design idea of the flexible seeding method in the main text.

7.) Page 5, L134: if you add ice number do you have to worry about conservation or inconsistencies? Not for number, but do such affects bleed into humans.

Reply: In the cirrus seeding experiments in this study, the seeding ice crystals are made from ambient atmospheric water vapor rather than artificially adding water into the atmosphere. Therefore, it is unnecessary to consider conservation or inconsistencies. We emphasized this in the experimental setups of the revised manuscript.

8.) Page 5, L140: does the ice added feedback through the microphysics?

Reply: Yes, it does. The seeding ice crystals not only impact the ice nucleation process but also impact other cloud microphysics processes because the ice crystals are directly added to the microphysics scheme. As shown in Section 3 of the manuscript, the impacts of cirrus seeding are very complicated. There is not only the direct instantaneous impact but also the feedback through microphysics.

9.) Page 10, L226: for clarity maybe you could pull out the global mean values into a table or a set of histograms.

Reply: Thank you for the comment. A new table was added in the revised manuscript as follows:

Table 2. Global annual mean cloud radiative effects from all experiments [a]. The corresponding standard deviations calculated from the difference of each year for 10 years are shown in brackets.

Experiments	REF	HET−REF	OPT−REF	INP20−REF	INP200−REF	R10−REF	GT−REF
$iCRE_{SW}$ (W m^{-2})	−5.30	3.39(0.03)	3.25(0.05)	0.38(0.07)	0.30(0.05)	2.81(0.05)	1.99(0.04)
$iCRE_{LW}$ (W m^{-2})	11.79	−6.84(0.04)	−6.29(0.07)	−0.83(0.10)	−0.31(0.08)	−5.40(0.07)	−4.33(0.06)
iCRE (W m^{-2})	6.49	−3.45(0.02)	−3.04(0.03)	−0.44(0.04)	−0.01(0.04)	−2.58(0.03)	−2.34(0.03)
Effectiveness (%)		56.19(0.70)	49.40(0.62)	6.69(1.45)	−2.22(1.32)	43.02(0.85)	39.01(0.95)
mCRE (W m^{-2})	−6.20	1.06(0.13)	1.09(0.11)	0.20(0.11)	0.15(0.13)	0.90(0.10)	0.81(0.12)
lCRE (W m^{-2})	−24.69	1.06(0.14)	0.94(0.11)	0.07(0.17)	−0.07(0.13)	0.62(0.13)	0.03(0.17)
CRE (W m^{-2})	−28.43	−1.98(0.26)	−1.36(0.18)	−0.27(0.26)	0.35(0.28)	−1.25(0.22)	−2.00(0.25)

[a] Shown are the ice cloud shortwave radiative effect ($iCRE_{SW}$), ice cloud longwave radiative effect ($iCRE_{LW}$), ice cloud radiative effect (iCRE), cirrus seeding effectiveness (Effectiveness), mixed-phase cloud radiative effect (mCRE), liquid cloud radiative effect (lCRE), and all cloud radiative effect (CRE).

10.) Page 10, L240: please name and make a formal equation for the seeding effectiveness.

Reply: Thanks. The cirrus seeding effectiveness ($-100 * |\Delta iCRE / iCRE|$) was first proposed in Gasparini et al. (2020), which was used to show what proportion of iCRE is eliminated by cirrus seeding. In the revised manuscript, the equation has been clarified.

11.) Page 12, L274: can you comment more on the mechanism: why does adding ICs impact mixed phase and liquid clouds? Is this a real effect or a feature of the model formulation?

Reply: As discussed above, the impact on liquid clouds seems to be a real effect, and the impact on mixed phase clouds might be an artifact of the bulk model physics. In the revised manuscript, further comments on the mechanism were given in Section 3 (experimental results), and the robustness of the mechanism was analyzed in the discussion section.

12.) Page 12, L283: can you explain in a sentence or so why N increases with decreasing R?

Reply: The large ice crystal (R is relatively larger) has a larger surface area, consuming water vapor via deposition growth more efficiently. For the same amount of ice crystals, it is easier for larger ice crystals to prevent homogeneous nucleation from happening.

13.) Page 12, L284: which experiments are "these"?

Reply: Thank you for this comment. "In these seeding experiments" refers to the OPT, R10 and GT experiments. In the revised manuscript, we have clarified this.

14.) Page 12, L285-7: Im lost here. I thought the point of seeding was not to reach Sihom? Please clarify.

Reply: Yes, the point of seeding was not to reach S_{ihom}. Notably, seeding occurs only where homogeneous nucleation would occur without seeding. This seeding strategy was clearly described in Section 2.3 (flexible seeding method).

15.) Page 14, L314: can you explain why? Does it have to do with liquid and mixed phase clouds?

Reply: As shown in Fig. S1, the cooling effect of cirrus seeding is more significant in the winter hemisphere. In low solar noon zenith angle regions, the warming effects from liquid clouds are pronounced because the water vapor is sufficient and convective activity is intense. Thus, avoiding seeding in low solar noon zenith angle regions (GT experiment in the manuscript) can obtain a stronger cooling effect.

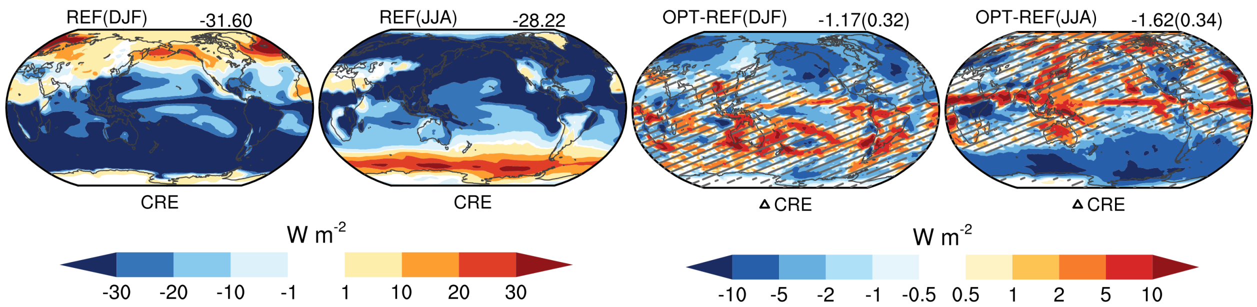

Figure S1: The annual mean spatial distribution of all cloud radiative effects (CREs) from the REF experiment for DJF and JJA (left panel) and the differences between the OPT and REF experiments (right panel). Global mean values are shown in the upper right corner, and the corresponding standard deviations calculated from the difference of each year for 10 years are shown in brackets.

16.) Page 14, L324: but don't you need INP to make IC?

Reply: It can be assumed that there is a machine that can make ice cubes from atmospheric water vapor and break the ice cubes into ice crystals.

17.) Page 15, L311: This is attributed to...

Reply: Thanks. Done.

18.) Page 15, L342: what is the mechanism for the effects on liquid and mixed phase clouds? You need to figure out of this is real or an artifact of the bulk model physics.

Reply: There might be many mechanisms for the effects on liquid and mixed-phase clouds. In the revised manuscript, the main possible mechanism was given. However, based on simulation results from one climate model, it is difficult to determine whether this is a real or an artifact of the bulk model physics. As discussed above, the comparison between our study and the study of Gasparini et al. (2017) suggests that the mechanism for the effect on liquid clouds might be a real physical mechanism, and the mechanism for the effects on mixed-phase clouds might be an artifact of the bulk model physics.

19.) Page 15, L353: is sedimentation the mechanism then? Can you test that?

Reply: The mechanisms for the warming effects from mixed-phase and liquid clouds are complicated. The increased convective activity might be the main mechanism, as discussed in Gasparini et al. (2017). Compared with the 50 μm ice crystal, the sedimentation of the 10 μm ice crystal is slower (i.e., longer lifetime in cirrus). In addition, N_{seedopt} from the R10 experiments (seeding ice crystal is 10 μm) is larger than that from the OPT experiment (seeding ice crystal is 50 μm). As a result, the decrease in the cirrus cloud radiative effect from the R10 experiments is weaker than that from the OPT experiment. Correspondingly, the increase in convective activity from the R10 experiment is weaker than that from the OPT experiment. To avoid misleading that sedimentation is the main mechanism of mixed-phase clouds formation, the sentence has been reworded.

20.) Page 17, L388: you say ICs increase W, but then contradict that in the next phase. This is confusing and unclear.

Reply: Thank you for this comment. Equation (A3) shows that the effect of pre-existing ICs (W_{pre}) and seeding ICs (W_{seed}) on ice nucleation can be taken as reducing the vertical velocity used for driving the ice nucleation parameterization. The effective updraft velocity (W_{eff}) used for ice nucleation is calculated as $W_{\mathrm{eff}} = W - W_{\mathrm{pre}} - W_{\mathrm{seed}}$, where W is the ambient updraft velocity. Increasing seeding ICs would increase the W_{seed}, and W would decrease.

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< In this revised manuscript, the main changes are marked in blue. >

Estimating the potential cooling effect of cirrus thinning achieved via the seeding approach

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Abstract. Cirrus thinning is a newly emerging geoengineering approach to mitigate global warming. To sufficiently exploit the potential cooling effect of cirrus thinning with the seeding approach, a flexible seeding method is used to calculate the optimal seeding number concentration, which is just enough to prevent homogeneous ice nucleation from occurring. A simulation using the Community Atmosphere Model version 5 (CAM5) with the flexible seeding method shows a global cooling effect of -1.36 ± 0.18 W m^{-2}, which is approximately two-thirds of that from artificially turning off homogeneous nucleation (-1.98 ± 0.26 W m^{-2}). However, simulations with fixed seeding ice nuclei particle number concentrations of 20 and 200 L^{-1} show a weak cooling effect of -0.27 ± 0.26 W m^{-2} and warming effect of 0.35 ± 0.28 W m^{-2}, respectively. Further analysis shows that cirrus seeding leads to a significant warming effect of liquid and mixed-phase clouds, which counteracts the cooling effect of cirrus clouds. This counteraction is more prominent at low latitudes and leads to a pronounced net warming effect over some low-latitude regions. The sensitivity experiment shows that cirrus seeding carried out at latitudes with solar noon zenith angles greater than 12° could yield a stronger global cooling effect of -2.00 ± 0.25 W m^{-2}. Overall, the potential cooling effect of cirrus thinning is considerable, and the flexible seeding method is essential.

1 Introduction

Global warming has been proven by observations and has been demonstrated many adverse effects on the environment and economy (Alexander et al., 2006; Feely et al., 2009; Lenton et al., 2019; Milne et al., 2009; Myhre et al., 2013). Saving energy and reducing greenhouse gas emissions are regarded as the primary strategies to counteract global warming, but these strategies may not be satisfactory (Fuss et al., 2018; IEA, 2019; Rogelj et al., 2015; Solomon et al., 2009). Therefore, geoengineering as a back-up tool against climate warming has been receiving increasing attention in recent years (e.g., Gasparini et al., 2020; Jones et al., 2018; Keith and MacMartin, 2015; Lawrence et al., 2018; Lohmann and Gasparini, 2017; Macnaghten and Owen, 2011). Geoengineering is usually divided into two categories: carbon dioxide removal (CDR), which aims to permanently eliminate CO_2 from the atmosphere, and solar radiation management (SRM), which proposes artificial intervention in the radiation budget (Caldeira et al., 2013; Heutel et al., 2018; Irvine et al., 2016; Kravitz et al., 2011; Vaughan and Lenton, 2011). It is well known that cirrus clouds (ice clouds) typically reflect less incoming solar radiation but block more of Earth's outgoing longwave radiation, which warms our planet (Berry and Mace, 2014; Hong et al., 2016; Matus and L'Ecuyer, 2017). Cirrus thinning geoengineering, which allows more longwave radiation to escape to space, leading to a cooling effect on the planet, has been investigated as a new SRM approach and has been proposed in Geoengineering Model Intercomparison Project Phase 6 (GeoMIP6, Kravitz et al., 2015). In GeoMIP6, cirrus thinning is simulated by artificially increasing the sedimentation velocity of ice crystals (ICs). Simulations with this idealized approach indicate that cirrus thinning can produce the desired globally averaged cooling effect (~ −2.0 W m^{-2}, e.g., Gasparini et al., 2017; Jackson et al., 2016; Muri et al., 2014). Considering the physical feasibility, simulating cirrus thinning by seeding with ice nuclei particles (INPs) is a better approach that can prevent homogeneous nucleation from occurring, thereby decreasing the number concentration of ICs (e.g., Gruber et al., 2019; Storelvmo and Herger, 2014; Storelvmo et al., 2013). Previous studies have shown that the cooling effect achieved via the seeding approach is sensitive to the seeding number concentration (N_{seed}), and even the strongest cooling effects are not strong enough (above −1.0 W m^{-2}, e.g., Gasparini and Lohmann, 2016; Gasparini et al., 2017; Penner et al., 2015). Note that the N_{seed} used in these model simulations is fixed (usually in the range of 0.1 to 200 L^{-1}), and the seeding strategy is uninterrupted (i.e., seeding occurs at every model time step). This study shows that the potential cooling effect of cirrus thinning cannot be sufficiently exploited due to the fixed seeding method. Moreover, a flexible seeding method is introduced to calculate the optimal N_{seed} (N_{seedopt}) based on the cirrus formation condition. The major purpose of this study is to estimate the potential cooling effect of cirrus thinning achieved via the seeding approach.

In this study, the cooling effects of cirrus thinning with different seeding methods are estimated. The paper is organized as follows. The flexible seeding method and its advantages are introduced in Sect. 2. This section also introduces the models and experimental designs that are employed. Comparisons of the cooling effects among different seeding methods and the main mechanism for the cooling effect are presented in Sect. 3. Finally, Sect. 4 presents the conclusions and discussion.

2 Methods and experiments

2.1 Cirrus thinning by seeding with ice nuclei particles

To better understand the seeding methods used in this study, it is necessary to briefly introduce the mechanism of cirrus thinning by seeding with INPs. In cirrus clouds, ICs are formed by homogeneous nucleation on soluble aerosol particles or heterogeneous nucleation on insoluble aerosol particles (Pruppacher and Klett, 1998). As ice-phase supersaturation (S_{i}) rises, heterogeneous nucleation occurs earlier with the aid of INPs (i.e., insoluble aerosols). A few ICs (usually less than 100 L^{-1}) are generated due to the relatively low number concentration of INPs. These newly formed ICs consume water vapor via deposition growth and then hinder S_{i} from rising (DeMott et al., 2003; Hoose and Möhler, 2012; Lohmann et al., 2008). The threshold S_{i} for homogeneous nucleation (S_{ihom}) is relatively higher. Therefore, homogeneous nucleation cannot occur (i.e., S_{i} cannot reach S_{ihom}) if there are enough newly formed ICs from heterogeneous nucleation. However, homogeneous nucleation can produce a large number of ICs once it takes place (usually much greater than 100 L^{-1}) because the number concentration of soluble aerosols in the upper troposphere is abundant (Barahona and Nenes, 2009; Kärcher, 2002). Therefore, seeding the clouds with a few INPs (usually less than 100 L^{-1}) can prevent S_{i} from reaching S_{ihom} (Barahona and Nenes, 2009; Liu and

Penner, 2005, McGraw et al., 2020). As a result, the in-cloud IC number concentrations (N_i) are usually decreased, which decreases the cirrus cloud optical depth (i.e., cirrus thinning; Storelvmo and Herger, 2014; Storelvmo et al., 2013).

2.2 Models and parameterizations

In this study, we use a cloud parcel model to illustrate the impact of seeding on the ice nucleation process. The parcel model presents the IC formation process in an adiabatically rising air parcel with a constant updraft vertical velocity (W). Equations that describe the evolution of temperature (T), pressure (P), ice water mixing ratio (Q_i), and ice particle size (R_i) can be found in Pruppacher and Klett (1998). The S_i is diagnosed from the conservation equation of total water (i.e., water vapor plus ice water). The homogeneous nucleation rate (J) of sulfate aerosol particles is calculated based on the water activity (Koop et al., 2000). The heterogeneous frozen fraction of dust aerosol particles is calculated by the classical nucleation theory (CNT, following Barahona and Nenes, 2009) with a freezing efficiency of 1.0 (i.e., 100% dust aerosols can act as INPs). More details about this cloud parcel model can be found in Shi and Liu (2016).

The climate model used in this study is version 5.3 of the Community Atmosphere Model (CAM5; Neale, 2012) with an improved ice nucleation parameterization that considers the effect of pre-existing ICs and in-cloud vertical velocity fluctuations (Shi et al., 2015; Shi and Liu, 2016). The ice nucleation parameterization considers the competition between heterogeneous freezing on coarse-mode dust aerosol particles and homogeneous freezing on sulfate aerosol particles. Here, 100% coarse-mode dust aerosols can act as INPs (Liu and Penner, 2005; Shi et al., 2015). Considering that sulfate aerosol particles may transform into glassy at very low temperatures (Murray et al., 2010), homogeneous nucleation is switched off below −68 °C (~205 K), which could make the modeled N_i to be close to observations (Shi et al., 2013). Note that there is no homogeneous nucleation in mixed-phase clouds (0°C ≥ T > −37 °C). The sub-grid vertical velocity (W_{sub}) derived from the turbulent kinetic energy is used to drive ice nucleation parameterization (Gettelman et al., 2010). The effect of pre-existing ICs on ice nucleation is parameterized by reducing the vertical velocity for ice nucleation (W_{pre}; Barahona et al., 2014; Kärcher et al., 2006; Shi et al., 2015). In the improved ice nucleation parameterization, the newly nucleated IC number concentration (N_{inuc}) is calculated as a function of T, P, S_i, W_{sub}, W_{pre}, the number concentration of coarse-mode dust aerosols (N_{dust}), and the number concentration of sulfate aerosols (N_{sul}). The cloud microphysics is represented by a two-moment scheme (Morrison and Gettelman, 2008).

2.3 Flexible seeding method

According to the mechanism of cirrus thinning caused by seeding with INPs, it is clear that $N_{seedopt}$ is the minimal number concentration to prevent homogeneous nucleation from occurring. If N_{seed} is less than $N_{seedopt}$ (underseeding), the newly formed ICs from heterogeneous nucleation are insufficient; thus, homogeneous nucleation still occurs and produces a relatively large N_{inuc}. If N_{seed} is larger than $N_{seedopt}$ (overseeding), despite homogeneous nucleation being suppressed, N_{inuc} remains somewhat larger due to excessive N_{seed}. Notably, in terms of consuming water vapor and hindering homogeneous nucleation, it is clear that ICs are superior to INPs (Pruppacher and Klett, 1998; Kärcher et al., 2006; Shi et al., 2015). In other words, ICs can act as cheaper, cleaner, and safer INPs. Therefore, ICs are used as the seeding material in the flexible seeding method introduced by this study. The formulas for calculating $N_{seedopt}$ are introduced in the Appendix. $N_{seedopt}$ is a function of cirrus ambient conditions, aerosol properties, and radius of seeding ICs (R_{seed}). R_{seed} is a tunable given parameter. It is important to point out that seeding with ICs occurs only where homogeneous nucleation would occur without seeding (i.e., flexible seeding strategy). The left panel in Fig. 1 illustrates the advantage of $N_{seedopt}$. Parcel model results show that without seeding (REF, black lines), heterogeneous nucleation takes place at S_i >10% and produces 10 L^{-1} of ICs. Because these newly formed ICs are too few to prevent S_i from increasing, homogeneous nucleation takes place at $S_i > S_{ihom}$ (~ 56%) and produces a large number of ICs (2937 L^{-1}). The final N_i (i.e., N_{inuc}) is 2947 L^{-1}. In the simulation with pure heterogeneous nucleation (HET, green lines), the final N_i is 10 L^{-1}. In the simulation that seeding with 28 L^{-1} ($N_{seedopt}$ is 28 L^{-1}) of ICs (OPT, red lines), the newly formed ICs from heterogeneous nucleation (10 L^{-1}) and seeding ICs are just enough to prevent S_i from reaching S_{ihom}. The final N_i (i.e., N_{inuc} + $N_{seedopt}$) is 38 L^{-1}. In the simulation that seeding with 20 L^{-1} of coarse-mode dust aerosol particles (INP20, blue lines, underseeding), heterogeneous nucleation produces more ICs (30 L^{-1}) than the REF simulation. However, homogeneous nucleation still occurs and produces 715 L^{-1} of ICs. The final N_i is 745 L^{-1}. In the simulation that seeding with 200 L^{-1} of coarse-mode dust aerosol particles (INP200, orange lines, overseeding), the newly formed ICs from heterogeneous nucleation (210 L^{-1}) are large enough to prevent homogeneous nucleation from occurring. The final N_i is 210 L^{-1}. Overall, seeding with INPs/ICs can lead to a lower N_i, and N_i from the OPT simulation is closest to the HET simulation.

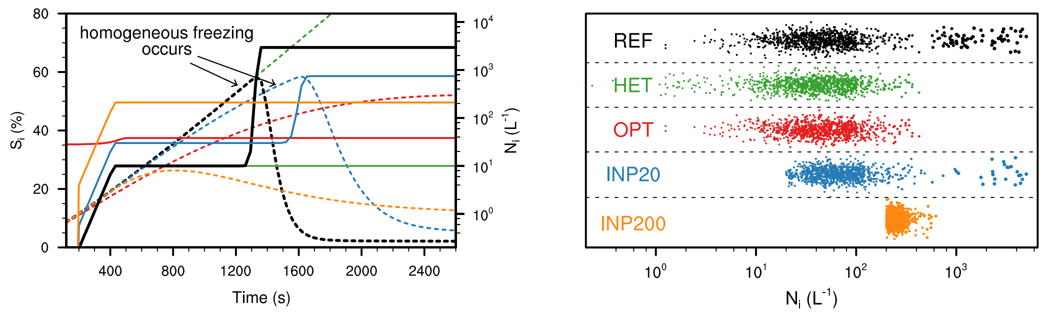

Figure 1: Schematic diagram of different seeding methods, reference results without seeding (REF, black), pure heterogeneous nucleation (HET, green), seeding with the optimal number concentration of ICs (OPT, red), seeding with 20 L^{-1} of INPs (INP20, blue), and seeding with 200 L^{-1} of INPs (INP200, orange). The optimal seeding method uses ICs with the radius of 25 µm. The left panel shows simulation results from the parcel model with given initial conditions (P = 330 hPa, T = 220 K, W = 0.3 m s^{-1}, N_{dust}=10 L^{-1}, and N_{sul} = 500 000 L^{-1}). The solid lines denote the total number concentrations of ICs in the parcel (N_i, units: L^{-1}), which include the seeding ICs and the newly formed ICs, and the dashed lines denote ice supersaturation (S_i, units: %). The arrows point to the beginning of homogeneous nucleation. The right panel shows the N_i from ice nucleation parameterizations driven by the same 1000 datasets of input variables (one dot denotes one offline result), which are sampled from the CAM5 simulation. The horizontal coordinate axis is N_i, and the vertical coordinate axis is meaningless.

Additionally, we run large-ensemble ice nucleation offline experiments to show the advantage of the flexible seeding strategy (Fig. 1, right panel). A total of 1000 cirrus formation cases are sampled from the CAM5 simulation without seeding. The input variables (T, P, S_i, W_{sub}, $N_{dust,}$ and N_{sul}) used to drive ice nucleation parameterization in CAM5 are used to drive these offline experiments. Homogeneous nucleation events account for 7.4 % (i.e., 74 homogeneous nucleation cases). Five experiments corresponding to the parcel model simulations are carried out. Each experiment is driven by the same 1000 cases. In the two fixed seeding experiments (i.e., the INP20 and INP200 experiments), INPs (i.e., coarse-mode dust aerosols) are added for all 1000 cases even if there were no homogeneous nucleations (i.e., uninterrupted seeding strategy). Compared with the REF experiment, all large N_i cases (dots with N_i > 500 L^{-1}) totally vanish in the HET experiment because only heterogeneous nucleation events occur. The N_i distribution in the OPT experiment is similar to that in the HET experiment except for some low N_i (< 10 L^{-1}) cases. In the INP20 experiment, there are some large N_i cases because homogeneous nucleation still occurs in 36 cases. In the INP200 experiment, there are no large N_i cases because almost all homogeneous nucleation cases are suppressed, whereas the N_i from all cases is greater than 200 L^{-1} due to the large N_{seed}. In short, the flexible seeding method is better than the fixed seeding method.

2.4 Experimental setups

CAM5 model experiments are carried out to estimate the cooling effect of cirrus thinning. Table 1 summarizes all the experiments performed in this study. The REF, HET, OPT, INP20, and INP200 experiments correspond to the offline experiments discussed above. In the INP20 and INP200 experiments, the N_{dust} used for driving ice nucleation parameterization (cirrus clouds only) increases by 20 and 200 L^{-1} (i.e., the N_{dust} from the aerosol module plus 20 and 200 L^{-1}), respectively. Note that the seeding INPs are added at every model time step but only impact the ice nucleation process (i.e., the N_{dust} in the aerosol module is not influenced by the seeding INPs). In the OPT experiment, the seeding ICs are directly added into the cloud microphysics scheme. As a result, these seeding ICs would affect both the ice nucleation process and other cloud microphysics processes. Notably, it is unnecessary to consider water conservation because the seeding ICs are made from ambient atmospheric water vapor.

Table 1. List of CAM5 experiments

Experiments	Description
REF	Reference experiment without cirrus thinning.
Cirrus thinning with different methods	
HET	Pure heterogeneous nucleation, homogeneous nucleation is artificially turned off.
OPT	Implement seeding globally using the flexible seeding method with IC radius (R_{seed}) of 50 μm.
INP20	Implement seeding globally with 20 L^{-1} of INPs.
INP200	Implement seeding globally with 200 L^{-1} of INPs.
Sensitivity experiments for the flexible seeding method	
R10	Similar to OPT, but R_{seed} is set to 10 μm.
GT	Similar to OPT, but seeding occurs over target regions, where the solar noon zenith angles are greater than 12°.

Additionally, we set up two sensitivity experiments for the flexible seeding method (Table 1). First, the tunable parameter R_{seed} is investigated. R_{seed} is 10 μm in the R10 experiment, whereas R_{seed} is 50 μm in the OPT experiment. Second, the seeding region is investigated. Cirrus thinning also leads to more incoming solar radiation (warming effect), which counteracts the cooling effect from more outgoing longwave radiation, especially for the low solar noon zenith angle regions (Storelvmo and Herger, 2014; Storelvmo et al., 2014). Furthermore, this study also finds that the cooling effect over low-latitude regions is less susceptible to cirrus seeding for other reasons (see Sect. 3.1). Thus, another sensitivity experiment with a specific geographical target (i.e., the GT experiment) is examined. Similar to the study of Storelvmo and Herger (2014), cirrus seeding is only carried out at latitudes where the solar noon zenith angles are greater than 12°, which compose approximately 80% of the Earth's surface.

In this study, all CAM5 experiments are atmosphere-only simulations with the same prescribed climatological ocean surface conditions. All experiments run for 11 model years at a horizontal resolution of 1.9°×2.5° and a model time step of 30 min. The first year is considered to be a spin-up period, and the last 10 years are used in the analyses. The standard deviations, which are estimated from the averages of each year, are used for variability analysis.

3 Estimating the cooling effect of cirrus thinning

3.1 Comparisons among different seeding methods

First, we analyze the impact of cirrus seeding on the ice nucleation process (Fig. 2). The contribution of homogeneous nucleation to cirrus formation (F_{hom}) is essential for the radiative properties of cirrus clouds (Jensen et al., 2013; Shi and Liu, 2016). Here, F_{hom} is quantified as the ratio of the homogeneous nucleation occurrence frequency to the ice nucleation occurrence frequency (F_{nuc}). In the REF experiment, F_{hom} is low near dust source regions (e.g., the Saharan Desert and Arabian Desert). F_{hom} is high over other tropical regions due to the large W_{sub} (not shown). Generally, F_{hom} is low (< 20%) in most regions, which is consistent with observations that heterogeneous nucleation is the dominant mechanism for cirrus formation (Cziczo et al., 2013; Jensen et al., 2013). Although F_{hom} from the INP20 experiment is decreased substantially, there are still some homogeneous freezing events (3.38% of all cirrus and 5.20% at 233 hPa). In the INP200 experiment, there are only a few homogeneous freezing events (0.42% of all cirrus and 0.63% at 233 hPa) due to the larger N_{seed} of INPs. Both the INP20 and INP200 experiments show that the averaged number concentration of ICs produced from heterogeneous freezing events (N_{ihet}) are increased. This increase would lead to more intense competition between homogeneous and heterogeneous nucleation. As a result, the averaged number concentrations of ICs produced from homogeneous freezing events (N_{ihom}) from the INP20 and INP200 experiments are substantially decreased compared with that from the REF experiment. As expected, F_{hom} and N_{ihom} are zero from the HET and OPT experiments. It is noteworthy that a large number of small ICs (e.g., homogeneous nucleation occurs) would exist for a long time, consuming water vapor via deposition growth and then hindering subsequent ice nucleation (Shi et al., 2015). Therefore, F_{nuc} from the REF experiment is very low (< 4%) in most regions, and F_{nuc} from the cirrus thinning experiments (i.e., the HET, OPT, INP20, and INP200 experiments) are obviously increased due to the decreases in F_{hom} and N_{ihom}. This finding suggests that the impact of cirrus seeding (including the HET experiment) on the ice nucleation process is very complicated. There is not only the direct instantaneous impact but also the indirect impact caused by subsequent changes.

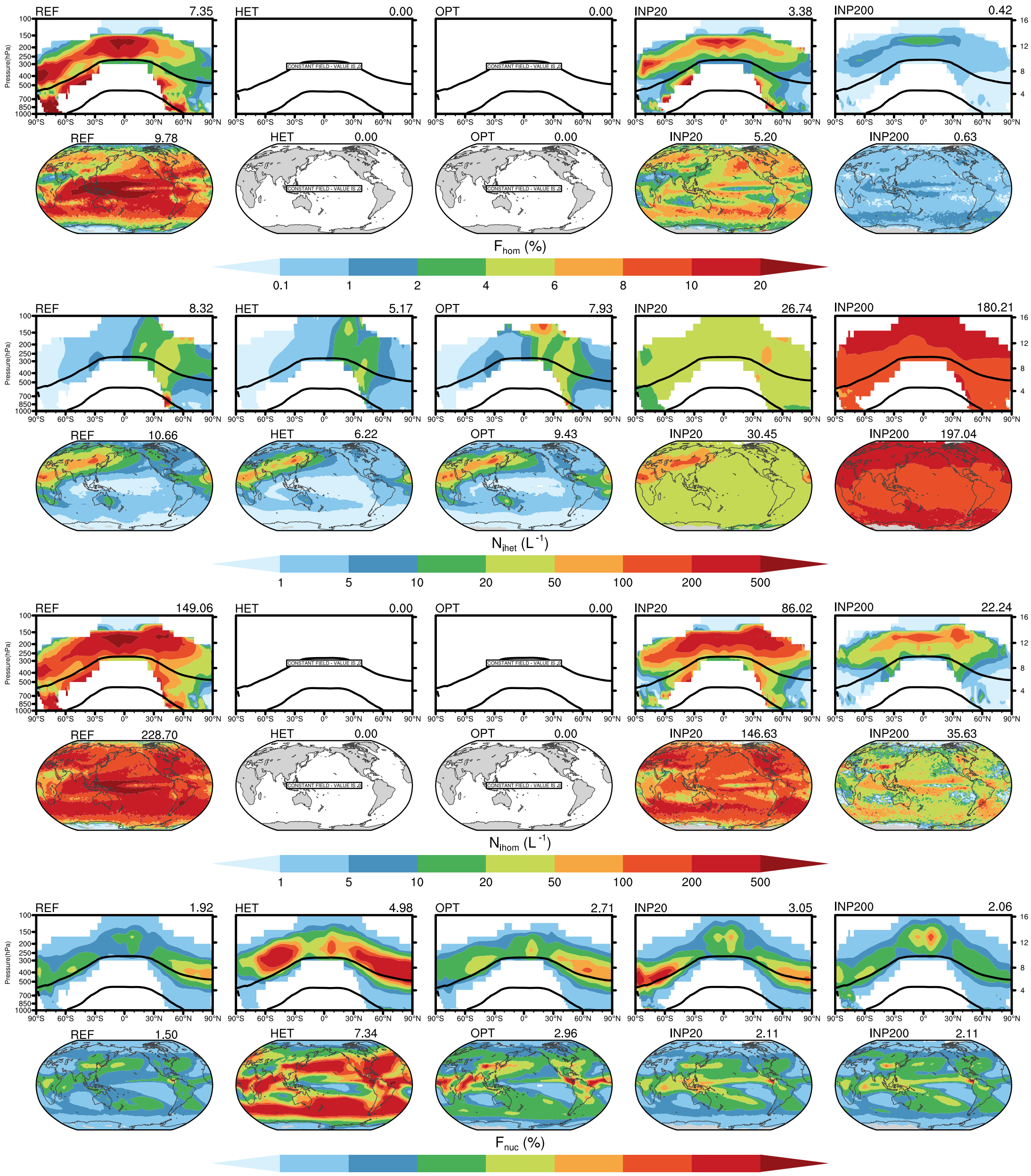

Figure 2: Annual zonal mean and 233 hPa spatial distribution of the homogeneous nucleation contribution to cirrus formation (F_{hom}, first panel), averaged IC number concentration produced from heterogeneous freezing events (N_{ihet}, second panel) and from homogeneous freezing events (N_{ihom}, third panel), and ice nucleation occurrence frequency (F_{nuc}, last panel). Experimental names are shown in the upper left corner, and global mean values are shown in the upper right corner. The two black lines are 0 and –37 °C isotherms. The results are sampled from model grids where F_{nuc} are greater than 0.1%.

Figure 3 shows the impact of cirrus seeding on cloud properties. In the cloud microphysics scheme, the in-cloud IC number concentration (i.e., N_i) mainly depends on the ice nucleation process (i.e., N_{inuc}, Shi et al., 2015; Shi and Liu, 2016). Therefore, the annual averaged N_i from the cirrus thinning experiments are decreased significantly in most cirrus clouds (ice clouds), especially from the HET and OPT experiments. The N_i from the INP20 experiment is not significantly decreased over the tropical regions because there are still many homogeneous freezing events (F_{hom} and N_{ihom} in Fig. 2). Compared with the REF experiment, N_i from the INP200 experiment is obviously increased in the tropical upper troposphere and the polar troposphere. This increase occurs because the homogeneous nucleation contribution (i.e., $F_{nuc} \times F_{hom} \times N_{ihom}$) from the REF experiment is relatively low, and the heterogeneous nucleation contribution (i.e., $F_{nuc} \times N_{ihet}$) from the INP200 experiment increases dozens of times over these regions (Fig. 2). Similarly, the vertically integrated N_i (i.e., column N_i) from the HET and OPT experiments are significantly decreased in most regions. In contrast, the changes in column N_i from the INP20 and INP200 experiments are not notable. The changes in ice water content (IWC) and ice water path (IWP) from the INP20 and INP200 experiments are also non-significant in most regions. In the HET and OPT experiments, both N_i and IWC in the middle and lower mixed-phase clouds are obviously increased. The main reason might be that the deep convective activity becomes more vigorous because cirrus thinning reduces atmospheric stability via the radiative budget (not shown). The ratio of ice to total cloud condensate detrained from the convective parameterizations is a linear function of temperature between −40 °C and −10 °C (Morrison and Gettelman, 2008). Furthermore, the ICs can grow through the Bergeron process in mixed-phase clouds (Morrison and Gettelman, 2008). This might be the reason that the relative increases in IWC in mixed-phase clouds are stronger than the relative increases in N_i. Although the increases in IWC in mixed-phase clouds counteract the decreases in IWC in ice clouds to some extent, the IWP are still significantly decreased in most regions from the HET and OPT experiments. However, the IWP are significantly increased over a few regions (e.g., middle Africa and northern Brazil) because the decreases in IWC in ice clouds are slight and even smaller than the increases in IWC in mixed-phase clouds over there (not shown). The changes in liquid water content (LWC) and liquid water path (LWP) from the INP20 and INP200 experiments are non-significant in most regions, whereas both the LWC and LWP from the HET and OPT experiments are significantly decreased in some low- and mid-latitude regions. One possible reason is that falling ICs accrete by riming of cloud droplets (Gasparini et al., 2017; Storelvmo et al., 2013), and the conversion efficiency of cloud droplets to precipitation is increased. Another possible reason is that cirrus thinning leads to stronger convective precipitation (Kristjánsson et al., 2015; Storelvmo and Herger, 2014), which would consume more cloud water (Gasparini et al., 2017; Rapp et al., 2011). The above analyses are in agreement with previous studies, which show that cirrus thinning might result in complex impacts on mixed-phase and liquid clouds (Gasparini and Lohmann, 2016; Gruber et al., 2019).

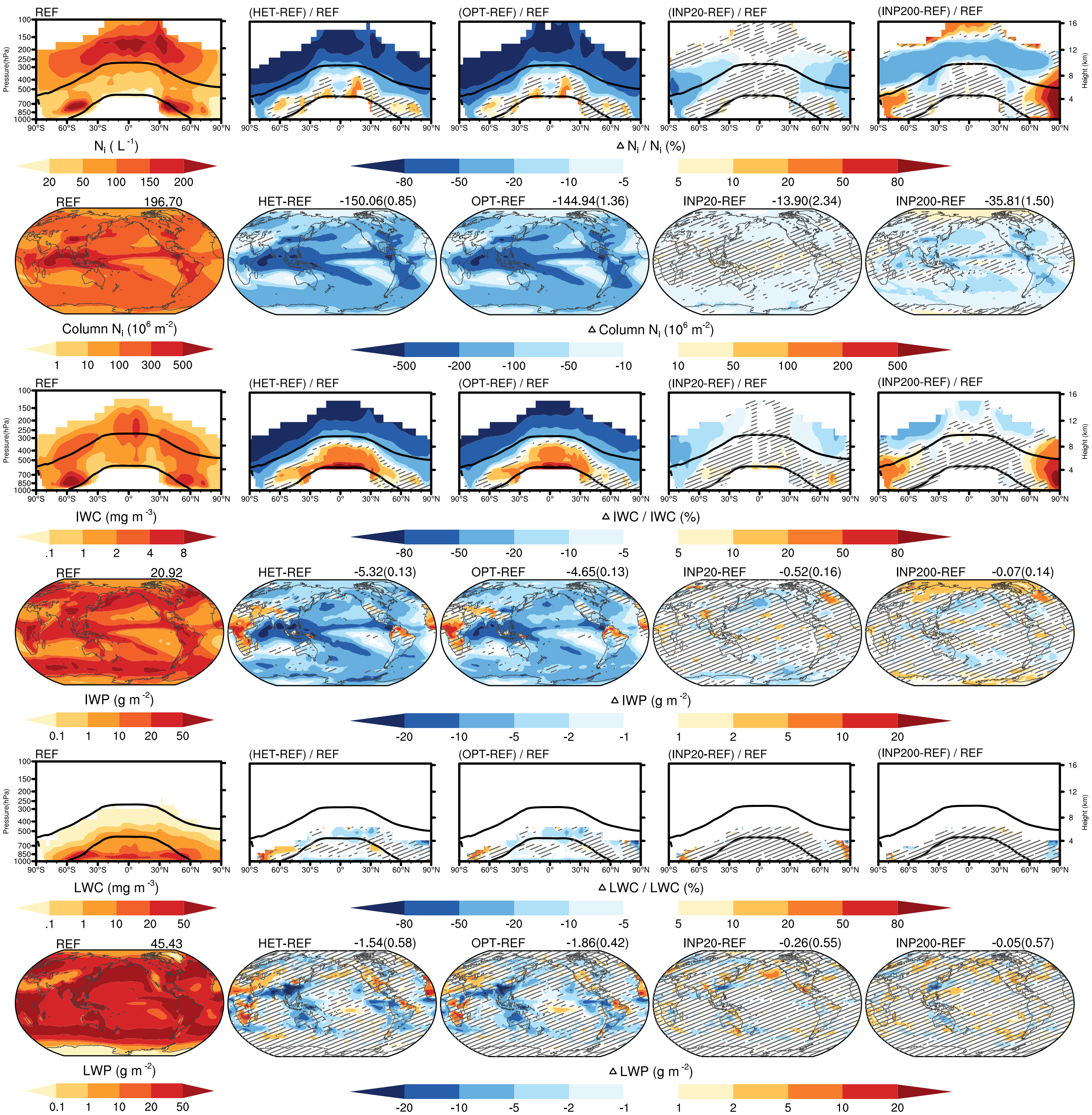

Figure 3: Annual zonal mean of in-cloud IC number concentration (N_i, first row), ice water content (IWC, third row), and liquid water content (LWC, fifth row) from the REF experiment (first column) and the relative changes from the HET, OPT, INP20 and INP200 experiments with respect to the REF experiment (second to fifth columns). The corresponding spatial distributions of vertically integrated N_i (Column N_i, second row), ice water path (IWP, fourth row), and liquid water path (LWP, sixth row) from the REF experiment and the differences ("Δ") from the HET, OPT, INP20 and INP200 experiments with respect to the REF experiment. Global mean values are shown in the upper right corner, and the standard deviations calculated from the difference of each year for 10 years are shown in brackets. The shadow denotes that the differences between two experiments are not significant at the 95% level based on Student's t-test.

The cooling effect of cirrus thinning is usually quantified by the anomaly in cloud radiative effect (ΔCRE; Mitchell and Finnegan, 2009; Storelvmo and Herger, 2014). For convenience of expression, "Δ" indicates the difference between the cirrus thinning experiments and the REF experiment. In addition to the model standard diagnostics of CRE, CRE from ice clouds (iCRE), mixed-phase clouds (mCRE), and liquid clouds (lCRE) are also diagnosed separately. Note that cirrus clouds are clouds at temperatures below –37 °C and above 440 hPa (Boucher et al., 2013), so we refer to them as ice clouds in this study. Furthermore, the cooling radiative effect is quantified by a negative value even if it has been declared a cooling effect. For convenience, the global annual mean cloud radiative effects from all experiments are listed in Table 2.

Table 2. Global annual mean cloud radiative effect from all experiments [a]. The corresponding standard deviations calculated from the difference of each year for 10 years are shown in brackets.

Experiments	REF	HET−REF	OPT−REF	INP20−REF	INP200−REF	R10−REF	GT−REF
$iCRE_{SW}$ (W m^{-2})	−5.30	3.39(0.03)	3.25(0.05)	0.38(0.07)	0.30(0.05)	2.81(0.05)	1.99(0.04)
$iCRE_{LW}$ (W m^{-2})	11.79	−6.84(0.04)	−6.29(0.07)	−0.83(0.10)	−0.31(0.08)	−5.40(0.07)	−4.33(0.06)
iCRE (W m^{-2})	6.49	−3.45(0.02)	−3.04(0.03)	−0.44(0.04)	−0.01(0.04)	−2.58(0.03)	−2.34(0.03)
Effectiveness (%)		56.19(0.70)	49.40(0.62)	6.69(1.45)	−2.22(1.32)	43.02(0.85)	39.01(0.95)
mCRE (W m^{-2})	−6.20	1.06(0.13)	1.09(0.11)	0.20(0.11)	0.15(0.13)	0.90(0.10)	0.81(0.12)
lCRE (W m^{-2})	−24.69	1.06(0.14)	0.94(0.11)	0.07(0.17)	−0.07(0.13)	0.62(0.13)	0.03(0.17)
CRE (W m^{-2})	−28.43	−1.98(0.26)	−1.36(0.18)	−0.27(0.26)	0.35(0.28)	−1.25(0.22)	−2.00(0.25)

[a] Shown are the ice cloud shortwave radiative effect ($iCRE_{SW}$), ice cloud longwave radiative effect ($iCRE_{LW}$), ice cloud radiative effect (iCRE), cirrus seeding effectiveness (Effectiveness), mixed-phase cloud radiative effect (mCRE), liquid cloud radiative effect (lCRE), and all cloud radiative effect (CRE).

The iCRE and its shortwave ($iCRE_{SW}$) and longwave ($iCRE_{LW}$) components are analyzed first (Fig. 4). The globally averaged iCRE from the REF experiment is 6.49 W m^{-2} (net warming effect) with a shortwave component ($iCRE_{SW}$) of −5.30 W m^{-2} (cooling effect) and a longwave component ($iCRE_{LW}$) of 11.79 W m^{-2} (stronger warming effect). This globally averaged iCRE is within the possible range reported in recent studies (4.5-6.8 W m^{-2}; Gasparini and Lohmann, 2016; Gasparini et al., 2020; Hong et al., 2016; Lohmann and Gasparini, 2017; Muench and Lohmann, 2020). The globally averaged $iCRE_{SW}$ from the HET, OPT, INP20, and INP200 experiments increase (less negative, warming effect) by 3.39, 3.25, 0.38, and 0.30 W m^{-2}, respectively. The decrease in $iCRE_{LW}$ (cooling effect) from all cirrus thinning experiments are stronger, especially from the HET (−6.84 W m^{-2}) and OPT (−6.29 W m^{-2}) experiments. Although $\Delta iCRE_{LW}$ from the HET and OPT experiments show significant cooling effects over most regions, there are still a few regions with warming effects (middle Africa and northern Brazil) due to higher ice cloud occurrence frequencies (not shown). The spatial patterns of $\Delta iCRE_{SW}$ and $\Delta iCRE_{LW}$ are generally in agreement with the changes in IWP and column N_i (Fig. 3). In terms of ΔiCRE, the HET (−3.45 W m^{-2}) and OPT (−3.04 W m^{-2}) experiments show much stronger cooling effects than the INP20 (−0.44 W m^{-2}) and INP200 (−0.01 W m^{-2}) experiments. Following Gasparini et al. (2020), the cirrus seeding effectiveness (−100 * |ΔiCRE / iCRE|) is used to show what proportion of iCRE is eliminated by cirrus seeding. The globally averaged cirrus seeding effectiveness from the HET and OPT experiments are 56.19% and 49.40%, respectively. These values are much higher than those from the INP20 (6.69%) and INP200 (−2.22%) experiments. The fixed seeding method restricts the cirrus seeding effectiveness. Notably, over some tropical regions, the cirrus seeding effectiveness from the HET and OPT experiments are somewhat low, although the ΔiCRE are relatively strong (< −5 W m^{-2}). One reason is that iCRE is relatively strong (> 10 W m^{-2}), but convective detrainment (anvil cirrus, which is not influenced by cirrus seeding) contributes more to iCRE (not shown). Another reason is that the ratio of Δ$iCRE_{SW}$ to Δ$iCRE_{LW}$ is higher over tropical areas due to the small solar noon zenith angles (not shown).

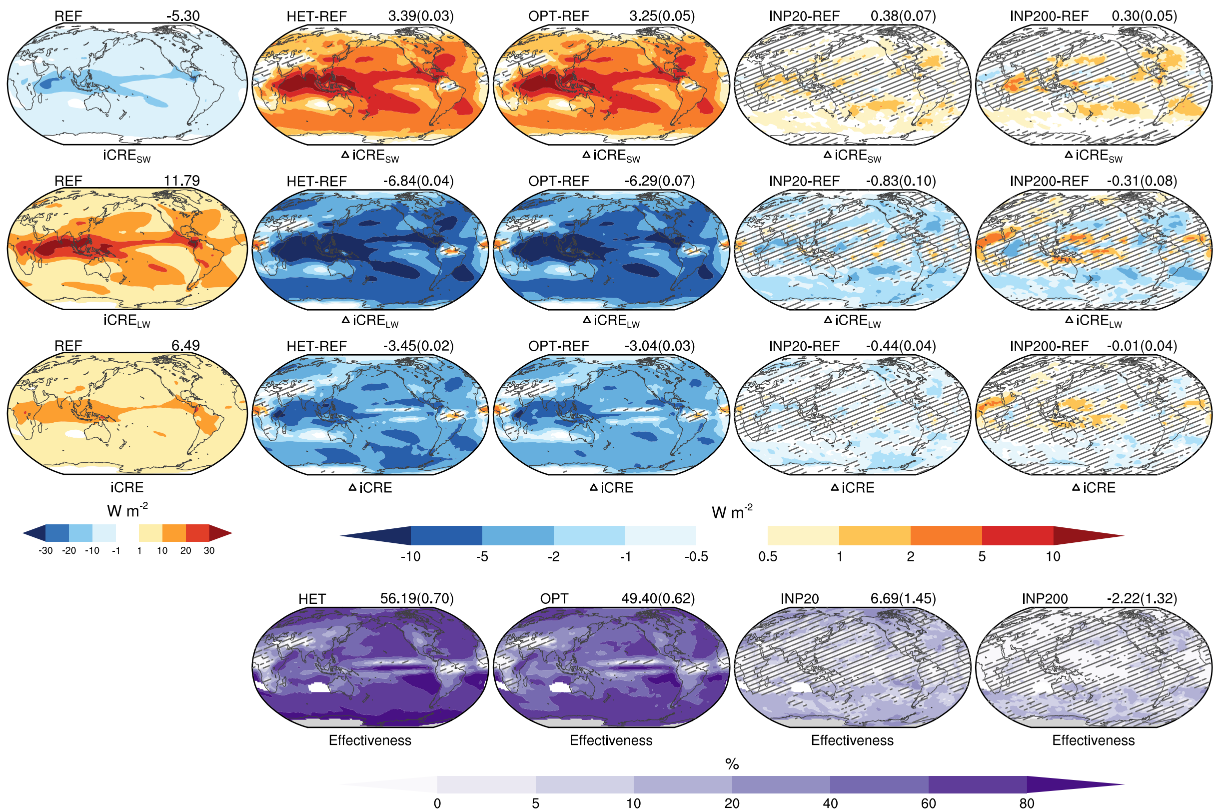

Figure 4: The annual mean spatial distribution of ice cloud shortwave radiative effect ($iCRE_{SW}$, first row), ice cloud longwave radiative effect ($iCRE_{LW}$, second row), ice cloud radiative effect (iCRE = $iCRE_{SW}$ + $iCRE_{LW}$, third row), and cirrus seeding effectiveness (fourth row) from the REF experiment (first column) and the differences ("Δ") from the HET, OPT, INP20 and INP200 experiments with respect to the REF experiment (second to fifth columns). Note that regions with absolute value of iCRE < 1.0 W m^{-2} from the REF experiment are excluded for calculating cirrus seeding effectiveness. Global mean values are shown in the upper right corner, and the corresponding standard deviations calculated from the difference of each year for 10 years are shown in brackets. The shadow denotes that the differences between two experiments are not significant at the 95% level based on Student's t-test.

In addition to iCRE, mCRE and lCRE are also obviously influenced by cirrus thinning (Fig. 5). Compared with the REF experiment, mCRE from the HET and OPT experiments are significantly increased in most ocean regions. The corresponding globally averaged ΔmCRE are 1.06 and 1.09 W m^{-2}, respectively. This warming effect (i.e., positive ΔmCRE) mainly comes from the increasing longwave component (not shown), which is consistent with the increase in IWC in mixed-phase clouds (Fig. 3). The globally averaged lCRE from the HET and OPT experiments increase (warming effect) by 1.06 and 0.94 W m^{-2}, respectively. The ΔlCRE is strong (> 2 W m^{-2}) over some low- and mid-latitude regions that couple with the decreases in LWP (Fig. 3). Both ΔmCRE and ΔlCRE from the HET and OPT experiments show that the globally averaged values are several times larger than the corresponding standard deviations (0.11-0.14). This finding indicates that cirrus thinning with the HET/OPT method leads to a significant globally averaged warming effect from mixed-phase clouds (ΔmCRE) and liquid clouds (ΔlCRE), although ΔmCRE and ΔlCRE are not statistically significant in most regions. Unlike the HET and OPT experiments, both ΔmCRE and ΔlCRE from the INP20 and INP200 experiments are weak and uncertain. The overall cooling effect of cirrus thinning (i.e., ΔCRE) from the HET and OPT experiments are −1.98 ± 0.26 W m^{-2} and −1.36 ± 0.18 W m^{-2}, respectively (Fig. 5). Compared with the cooling effect of ice clouds (i.e., ΔiCRE, Fig. 4), these values drop by approximately half due to the warming effect exerted by mixed-phase and liquid clouds. The INP20 and INP200 experiments show a weak cooling effect (−0.27 ± 0.26 W m^{-2}) and even a small warming effect (0.35 ± 0.28 W m^{-2}), respectively. It is clear that cirrus seeding with the flexible method could produce a notable global cooling effect, which is much better than the fixed methods. Furthermore, the cooling effect with the flexible seeding method is significant over most mid- and high-latitude regions. Some low-latitude regions show a pronounced warming effect because cirrus seeding leads to a stronger warming effect introduced by mixed-phase and liquid clouds (i.e., ΔmCRE and ΔlCRE). This finding suggests that cirrus seeding over low-latitude regions might be redundant.

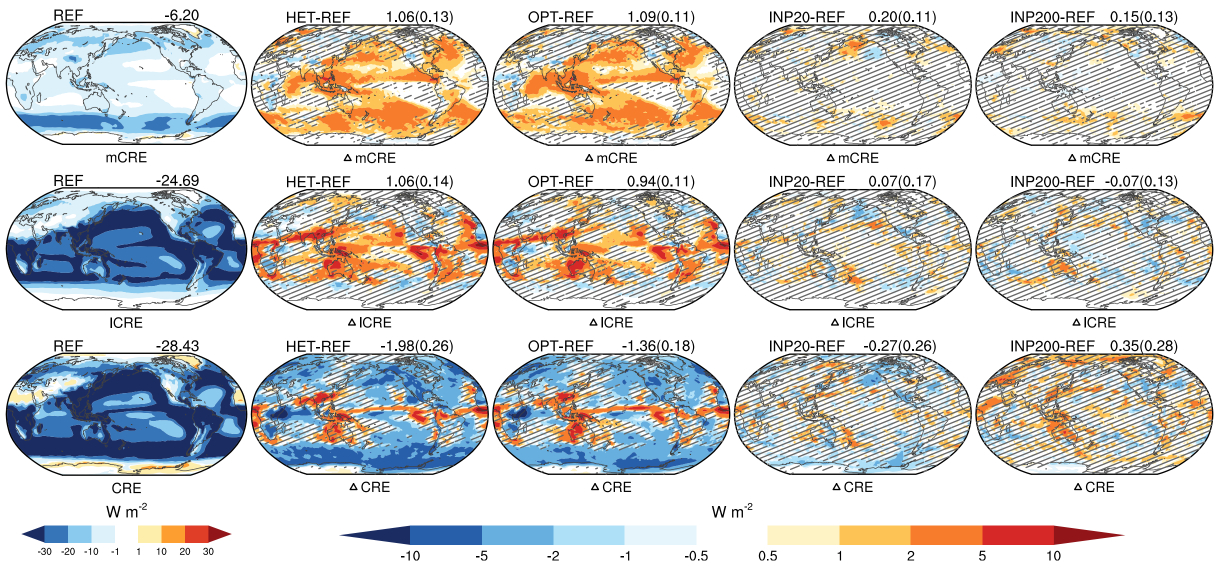

Figure 5: Similar to Fig. 4 but for the mixed-phase cloud radiative effect (mCRE, first row), liquid cloud radiative effect (lCRE, second row), and all cloud radiative effect (CRE, third row).

3.2 Sensitivity experiments with the flexible seeding method

To better understand cirrus thinning with the flexible seeding method, this section investigates sensitivity experiments of the cooling effect on R_{seed} (R10 experiment) and the seeding region (GT experiment).

Figure 6 shows the seeding number concentration ($N_{seedopt}$) and seeding frequency (F_{seed}). As expected, the OPT and GT experiments show similar $N_{seedopt}$ in mid- and high-latitude regions. In these two experiments, $N_{seedopt}$ is less than 40 L^{-1} in most regions. Because $N_{seedopt}$ increases with decreasing R_{seed} (see Appendix), $N_{seedopt}$ from the R10 experiment is larger than that from the OPT and GT experiments. In these seeding experiments (i.e., the OPT, R10 and GT experiments), it becomes easier for the ice nucleation process to reach S_{ihom} (i.e., cirrus seeding occurs) because the large amount of long-lived small ICs produced by homogeneous nucleation is cut off. As a result, F_{seed} from the seeding experiments are much larger than the homogeneous freezing occurrence frequency ($F_{hom} \times F_{nuc}$) from the REF experiment (much less than 1%, Fig. 2). However, F_{seed} from the seeding experiments is still relatively low (< 4%) in most regions. F_{seed} from the GT experiment is even lower than 2% in most regions. The smaller ICs usually have a longer lifetime in cirrus clouds, so F_{seed} from the R10 experiment (1.01% of all cirrus and 1.05% at 233 hPa) is lower than that from the OPT experiment (1.13% of all cirrus and 1.43% at 233 hPa). Similar to the spatial distribution of F_{hom} from the REF experiment, F_{seed} from the cirrus seeding experiments are much higher in the low-latitude regions. This is the reason why the globally averaged F_{seed} from the GT experiment (0.82% of all cirrus and 0.80% at 233 hPa) is about one-third lower than that from the OPT experiment.

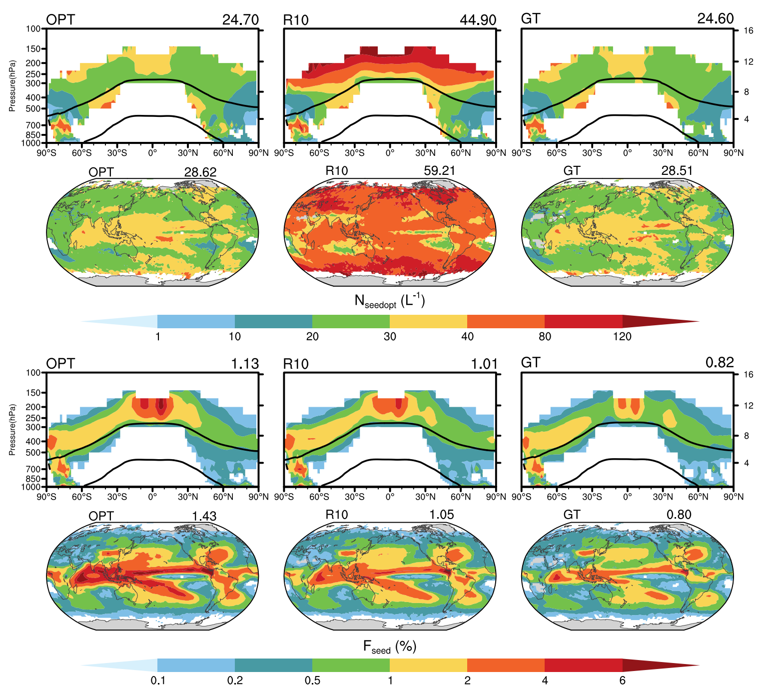

Figure 6: Annual zonal mean and 233 hPa spatial distribution of the optimal seeding number concentration ($N_{seedopt}$, first panel) and seeding frequency (F_{seed}, second panel). The names of the experiments are shown in the upper left corner, and globally mean values are shown in the upper right corner. The results are sampled from model grids where F_{seed} are greater than 0.1%.

Figure 7 shows the cooling effects from the R10 and GT experiments. The globally averaged ΔiCRE from the R10 experiment is −2.58 W m^{-2}. This ice cloud cooling effect is obviously weaker than that from the OPT experiment (−3.04 W m^{-2}) because the seeding ICs in the R10 experiment (larger $N_{seedopt}$ and smaller R_{seed}) could exist for a longer time in cirrus clouds. Correspondingly, the cirrus seeding effectiveness from the R10 experiment (43.02%) is also less than that from the OPT experiment (49.40%). Similar to the OPT experiment, the R10 experiment also shows that cirrus seeding induces an obvious global warming effect of mixed-phase and liquid clouds (ΔmCRE and ΔlCRE, Table 2). Notably, these warming effects (i.e., ΔmCRE and ΔlCRE) are weaker than those from the OPT experiments (Table 2). Thus, ΔCRE from the R10 experiment is −1.25 ± 0.22 W m^{-2}, which is close to that from the OPT experiment (−1.36 ± 0.18 W m^{-2}). Compared with the OPT experiment, ΔiCRE from the GT experiment becomes weaker over the regions without seeding (Figs. 7 and 4). Thus, the globally averaged ΔiCRE only decreases by −2.34 W m^{-2} from the GT experiment. Correspondingly, the cirrus seeding effectiveness from the GT experiment is also obviously less than that from the OPT experiment except in high-latitude regions. As mentioned in Sect. 3.1, cirrus seeding would lead to a strong warming effect of mixed-phase and liquid clouds at low latitudes. As expected, in the GT experiment, this warming effect is constrained to some extent (Table 2). The globally averaged cooling effect (ΔCRE) from the GT experiment is −2.00 ± 0.25 W m^{-2}, which is much stronger than that from the OPT experiment (−1.36 ± 0.18 W m^{-2}) and even stronger than that from the HET experiment (−1.98 ± 0.26 W m^{-2}). This finding suggests that cirrus seeding without low solar noon zenith angle regions might produce a better global cooling effect.

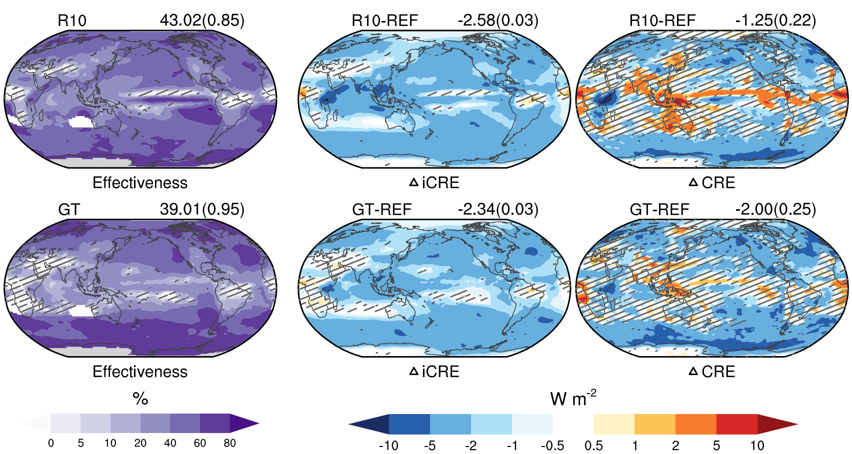

Figure 7: Similar to Fig. 4 but for ΔiCRE (middle), ΔCRE (right), and cirrus seeding effectiveness (left) from the R10 (upper panel) and GT (lower panel) experiments.

4 Conclusions and discussion

The major purpose of this study is to estimate the potential cooling effect of cirrus thinning. Based on the mechanism of cirrus thinning by the seeding approach, a flexible seeding method is used to calculate the optimal seeding number concentration, which is just enough to prevent homogeneous ice nucleation from happening. Furthermore, the cirrus seeding approach could move further by injecting ice crystals (ICs) instead of ice nuclei particles (INPs). In terms of hindering homogeneous nucleation and environmental safety, ICs are better than INPs. More importantly, the problem of INP transportation discussed in previous studies might be solved because ICs can be made from ambient atmospheric water vapor.

Both parcel model simulations and large-ensemble ice nucleation offline experiments show that the flexible seeding method has obvious advantages over the fixed seeding method. Furthermore, the CAM5 simulations with the flexible seeding method (implementing seeding globally) show a notable global cooling effect, -1.36 ± 0.18 W m^{-2} from seeding with ICs of 50 μm (OPT experiment) and -1.25 ± 0.22 W m^{-2} from seeding with ICs of 10 μm (R10 experiment). However, simulations with fixed seeding number concentrations of 20 and 200 INPs L^{-1} show a weak cooling effect of -0.27 ± 0.26 W m^{-2} and a warming effect of 0.35 ± 0.28 W m^{-2}, respectively. Note that some previous work using CAM5 with the fixed seeding method showed notable cooling effect (~ -2 W m^{-2}; e.g., Storelvmo and Herger, 2014; Storelvmo et al., 2014). This is attributed to the contribution of homogeneous nucleation to cirrus formation (F_{hom}) from the default CAM5 model used in their study (Penner et al., 2015). The F_{hom} from default CAM5 simulations is relatively higher because the default version neglects the effect of pre-existing ICs (Shi et al., 2015). Penner et al. (2015) tuned the main ice nucleation mechanism in CAM5 to limit F_{hom} and found that cirrus thinning with a fixed seeding number concentration cannot produce a definite global cooling effect. In this study, F_{hom} is also limited to a low level (Fig. 2). Our results with the fixed seeding method are similar to the study of Penner et al. (2015). However, with the benefits of the flexible seeding method, cirrus seeding could produce a considerable cooling effect.

This study also analyses the main mechanism for the cooling effect achieved via cirrus seeding. Simulation results show that cirrus seeding not only impacts ice clouds but also significantly impacts mixed-phase and liquid clouds. In terms of ice clouds, cirrus thinning with the flexible seeding method could lead to a notable cooling effect. However, cirrus seeding also leads to a significant warming effect of mixed-phase and liquid clouds, which counteracts the cooling effect of cirrus clouds. Because the counteraction is more prominent over low-latitude regions, the low-latitude regions are less susceptible to cirrus seeding. This finding agrees with the previous finding that cirrus thinning is more effective at mid and high latitudes because of more insolation caused by cirrus thinning when the sun is overhead (Storelvmo et al., 2014). The warming effect of liquid clouds from the OPT experiment (0.94 ± 0.11 W m^{-2}) is similar to the study of Gasparini et al. (2017; Table 5, 0.96 ± 0.25 W m^{-2} from the ECHAM-HAM model simulation that seeding with 1 L^{-1} of 50 μm INPs). There seems to be a relatively solid mechanism that cirrus thinning reduces atmospheric stability, leading to the warming effect of liquid clouds. However, the warming effect of mixed-phase clouds from the OPT experiment (1.09 ± 0.11 W m^{-2}) is several times stronger than that reported in their results (0.15 ± 0.10 W m^{-2}). This difference suggests that the climatic response to cirrus seeding is complex and might differ among different climate models and seeding methods. Finally, it is necessary to point out that the compensating effects introduced in this study (i.e., the warming effect of mixed-phase and liquid clouds) are derived from the atmosphere-only simulations with prescribed ocean surface conditions, the coupled model simulations might show different results (e.g., Gasparini et al., 2017).

Sensitivity experiments with the flexible seeding method show that smaller seeding ICs leads to a weaker global cooling effect of ice clouds due to the larger seeding number concentration and smaller ICs. The warming effects of mixed-phase and liquid clouds are also reduced to some extent because the convective activity from the R10 experiment is not as strong as that from the OPT experiment (not shown). Thus, the global cooling effect from seeding with smaller ICs (-1.25 ± 0.22 W m^{-2}) is not obviously weaker than seeding with larger ICs (-1.36 ± 0.18 W m^{-2}). Avoiding seeding over low-latitude regions can limit some warming effects due to changes in mixed-phase and liquid clouds and thus lead to a more pronounced global cooling effect. Sensitivity experiment shows that seeding carried out at latitudes with solar noon zenith angles greater than 12° yields a stronger global cooling effect of -2.00 ± 0.25 W m^{-2}, which is close to that of artificially turning off homogeneous nucleation over the whole Earth (-1.98 ± 0.26 W m^{-2}). In addition, we carried out sensitivity experiments with other threshold values (23.5°, 18°, and 8°). With increasing thresholds, the global cooling effect of ice clouds decreases, and the global warming effects of mixed-phase and liquid clouds also decrease. The overall cooling effect is maximized when using a solar zenith angle threshold of 12°. In short, the global cooling effect is more sensitive to seeding regions than to the radius of seeding ICs. The global cooling effect can thus be maximized when limiting seeding to the most suitable regions and times of the year. However, estimating the cooling effect of cirrus seeding based on commercial airliners (i.e., the limited time and place) is more realistic. We plan to investigate this method in our future work.

Appendix: The formula of optimal seeding number concentration (N_{seedopt})

For the ice nucleation parameterization with the pre-existing IC effect, the seeding ICs are considered to be pre-existing ICs. The optimal number concentration of ICs (N_{seedopt}) depends on the ice nucleation parameterization, especially for its treatment of the pre-existing IC effect.

Without the pre-existing ICs or seeding ICs, the temporal evolution of S_i is governed by the following (Kärcher et al., 2006):

$$\frac{\mathrm{d}S_\mathrm{i}}{\mathrm{d}t} = a_1 S_\mathrm{i} W - (a_2 + a_3 S_\mathrm{i}) \frac{\mathrm{d}Q_\mathrm{nuc}}{\mathrm{d}t}, \tag{A1}$$

where the parameters a_1, a_2, and a_3 only depend on the ambient temperature and pressure. W is the updraft velocity, and $\frac{\mathrm{d}Q_\mathrm{nuc}}{\mathrm{d}t}$ denotes the growth rate of newly nucleated ICs. To account for the effect of pre-existing ICs and seeding ICs, the deposition growth of pre-existing ICs ($\frac{\mathrm{d}Q_\mathrm{pre}}{\mathrm{d}t}$) and seeding ICs ($\frac{\mathrm{d}Q_\mathrm{seed}}{\mathrm{d}t}$) are added in Eq. (A1):

$$\frac{\mathrm{d}S_\mathrm{i}}{\mathrm{d}t} = a_1 S_\mathrm{i} W - (a_2 + a_3 S_\mathrm{i})\left(\frac{\mathrm{d}Q_\mathrm{nuc}}{\mathrm{d}t} + \frac{\mathrm{d}Q_\mathrm{pre}}{\mathrm{d}t} + \frac{\mathrm{d}Q_\mathrm{seed}}{\mathrm{d}t}\right), \tag{A2}$$

Equation (A2) can be rewritten as the following form:

$$\frac{\mathrm{d}S_\mathrm{i}}{\mathrm{d}t} = a_1 S_\mathrm{i}\left(W - W_\mathrm{pre} - W_\mathrm{seed}\right) - (a_2 + a_3 S_\mathrm{i}) \frac{\mathrm{d}Q_\mathrm{nuc}}{\mathrm{d}t}, \tag{A3}$$

$$W_\mathrm{pre} = \frac{a_2 + a_3 S_\mathrm{i}}{a_1 S_\mathrm{i}} \frac{\mathrm{d}Q_\mathrm{nuc}}{\mathrm{d}t}, \tag{A4}$$

$$W_\mathrm{seed} = \frac{a_2 + a_3 S_\mathrm{i}}{a_1 S_\mathrm{i}} \frac{\mathrm{d}Q_\mathrm{seed}}{\mathrm{d}t}, \tag{A5}$$

The effect of pre-existing ICs on ice nucleation can be taken as reducing the vertical velocity (W_pre, Barahona et al., 2014). Details about how to calculate W_pre are introduced in Shi et al. (2015). Here, the reduced vertical velocity from seeding ice (i.e., W_{seed}) is similar to W_{pre}. W_{seed} is a function of seeding ice number concentration (N_{seed}) and its radius (R_{seed}). Assuming all seeding ICs have the same R_{seed}, the growth rate is given by:

$$\frac{\mathrm{d}Q_{\mathrm{seed}}}{\mathrm{d}t} = \frac{4\pi\rho_i}{m_w} N_{\mathrm{seed}} \frac{b_1 R_{\mathrm{seed}}^2}{1+b_2 R_{\mathrm{seed}}}, \tag{A6}$$

where ρ_i is the ice density and m_w is the mass of a water molecule. $b_1 = \alpha v_{th} n_{sat} (S_i - 1)/4$, $b_2 = \alpha v_{th} n_{sat}/4D$. α is the water vapor deposition coefficient on ice, v_{th} is the thermal speed, n_{sat} is the water vapor number density at ice saturation, and D is the water vapor diffusion coefficient from the gas phase to the ice phase (Kärcher et al., 2006).

Under a given R_{seed}, W_{seed} increases with increasing N_{seed}. That is, the more ICs that are added, the more they will reduce W. The minimal N_{seed} (i.e., $N_{seedopt}$) is calculated based on the minimal W_{seed}, which can prevent homogeneous ice nucleation from occurring. The default ice nucleation parameterization (Liu and Penner, 2005; LP parameterization) provides a threshold updraft velocity (W_{thre}) for homogeneous ice nucleation,

$$W_{\mathrm{thre}} = e^{\frac{T-b}{a}}, \tag{A7}$$

where T is the ambient temperature, $a = -1.4938\ln N_{INP} + 12.884$, $b = -10.41\ln N_{INP} - 67.69$. N_{INP} is the INP (e.g., dust aerosol particle) number concentration. Homogeneous ice nucleation does not occur (i.e., only heterogeneous nucleation) if the effective updraft velocity (W_{eff}, $W_{eff} = W - W_{pre} - W_{seed}$) is less than W_{thre}. Thus, the minimal W_{seed} is calculated as $W_{seed} = W - W_{pre} - W_{thre}$. If $W_{seed} < 0$, there is no need for seeding. The minimal number concentration of seeding ICs (i.e., $N_{seedopt}$) can be calculated based on Eq. (A5) and (A6) at threshold S_i for homogeneous freezing (S_{ihom}). In this study, with the given R_{seed}, $N_{seedopt}$ is given by:

$$N_{\mathrm{seedopt}} = \frac{a_1 S_{\mathrm{ihom}} m_{\mathrm{w}}}{(a_2 + a_3 S_{\mathrm{ihom}})4\pi\rho_{\mathrm{i}}} \frac{1+b_2 R_{\mathrm{seed}}}{b_1 R_{\mathrm{seed}}^2} (W - W_{\mathrm{pre}} - W_{\mathrm{thre}}). \tag{A8}$$

Because the impact of deposition growth on pre-existing ICs is neglected in calculating W_{pre} (Barahona et al., 2014; Shi et al., 2015), the increase in R_{seed} caused by deposition growth during the ice nucleation process is also neglected. As a result, $N_{seedopt}$ might be overestimated, especially for a small given R_{seed}. The LP parameterization provides a critical number concentration of INPs (N_{lim}) for the only heterogeneous freezing scenario. $N_{seedopt}$ cannot exceed N_{lim} because ICs are superior to INPs for hindering homogeneous nucleation.

Code and data availability

The modified code of CAM5 and the output data used in this study are available online at DOI: 10.5281/zenodo.4507001.

Competing interests

The authors declare that they have no conflicts of interest.

Author contribution

Jiaojiao derived the formula for optimal seeding number concentrations. Xiangjun and Jiaojiao designed the model experiments and developed the model code. Jiaojiao processed and analyzed the raw model output data and wrote the paper. Xiangjun helped to explain the results. Both authors contributed to improving and reviewing the manuscript.

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