Does prognostic seeding along flight tracks produce the desired effects of cirrus cloud thinning? (egusphere-2022-1238) *Colin Tully, David Neubauer, Diego Villanueva, and Ulrike Lohmann* 28th March 2023

Referee #1 Author Response

To Referee #1,

Thank you for taking the time to read and review our manuscript.

I quoted each of your comments below with our responses and changes in the text where applicable.

Sincerely,

Colin Tully (on behalf of all co-authors)

Specific Comments

- 1. **Comment:** To my feeling, section 3.1 needs more structure so that the important results are easier to locate. Below I suggest some sub-sections where I think it is helpful.
 - a. **Response:** Thank you. We have addressed your specific comments below.
- Comment: Line 284ff: As expected, we find the largest positive net TOA anomaly when seeding with the largest average seeding particle number concentration (> 10⁵ L⁻¹, Fig. 2) that is associated with the case with a mean emissions radius of 0.01 μm (r0.01) and a mass scaling factor of 1000 (high-seeding). Why is it expected that a large number of seeding INPs result in a large positive net TOA?
 - a. **Response:** This specific case that you quote in the comment is the largest number of INPs we simulated in our study. As this represents the largest perturbation to the unseeded reference case it is expected that we find the largest net TOA anomaly. However, that could be either positive or negative. In the revised text, we cut "As expected,".

And why does the size matter? For example, the INP concentration from r0.1 and x1000 is almost identical with r0.01 and x10 /Fig. 4.2, but without a response (Fig. 4.3). Why is that?

a. **Response:** Aerosol size matters in our study as it impacts the rate of removal in the atmosphere, i.e. smaller particles will have a longer atmospheric lifetime and therefore their concentrations accumulate over time.

b. While the two cases you state above do have very similar INP concentrations, the r0.01x10 concentration is slightly higher on the log-scale in Figure 2 towards higher latitudes. We suspect this is why it shows a larger response than r0.1x1000.

In nature, we know that mostly larger INPS (> 0.5 μ m) will be activated, how does that correspond with your finding?

- a. **Response:** Our inclusion of smaller particles is justified based on previous work on the ice nucleation ability of AgI. We included some examples of this work in the revised text under the Experimental Setup section.
- b. Changes in the text:

"Including such small seeding particles in our model (0.01 μ m) is justified based on previous work on the ice nucleation ability of silve-iodide (AgI). Xue et al. (2013) formulated a parameterization for glaciogenic cloud seeding with AgI in the Weather Research and Forecasting (WRF) model, using a mean particle diameter of 0.04 μ m They reported that the model could reasonably produce the physical processes of cloud seeding. Geresdi et al. (2020) also investigated cloud seeding in the WRF model with slightly larger AgI with a mean diameter of 0.05 μ m and reported that the model also reasonably reproduced the microphysical properties of real clouds. Marcolli et al. (2016) reviewed lab-based experiments of ice nucleation and showed that AgI particles of 20 nm in diameter had an increasing ice nucleation efficiency towards cirrus temperatures (238 K). Finally, Kanji et al. (2017) presented new evidence of the ice nucleation ability of small particles such as pollen and fungal spores, which challenges arguments that only large particles are suitable INPs."

If it is explained later in the paper what causes the radiation feedbacks, please note that here (cross reference).

- c. **Response:** Agreed. We added a reference to the discussion section in the text.
- d. Changes in the text:

"To some extent this could be linked to optically thicker cirrus (Krämer et al., 2020) or optically thicker lower-lying mixed-phase or liquid clouds (Twomey, 1959, 1977; Albrecht, 1989), which is discussed further in Section 3.3."

3. **Comment:** Line 286 f: The large TOA anomalies are driven by a large increase in the LW cloud radiative effect (CRE) by 10.1 Wm⁻² (Tab. 3), indicating a significant change in cirrus cloud properties.

Please specify 'properties' (see also comment to Table 3).

a. **Response:** See response under your **Comment 27** for Table 3. We agree that this could be expanded, but only slightly. Therefore, we

amended this sentence in the revised text to point the reader to Figure 4 that shows that cloud fraction and increased ICNC are linked to the positive TOA anomalies we find.

b. Changes in the text:

"The large TOA anomalies are driven by a large increase in the LW cloud radiative effect (CRE) by 10.1 Wm⁻² (Tab. 3), indicating a significant change in cirrus cloud properties such as cloud fraction and ICNC (Fig. 4)."

4. **Comment:** Line 321 - the following paragraph:

This paragraph could be a sub-section with the title , Ice crystal sizes '

Question on the paragraph: Why you discuss here in detail the size anomaly? Is this because the size is one parameter influencing the radiative feedback? Because less sedimentation of smaller ice particles keeps the cirrus at higher altitudes (--> more warming)? Please explain.

- a. **Response:** We agree with the addition of a sub-section to 3.1 and added this into the revised manuscript.
- b. **Response to question:** Yes, exactly as you wrote, we discuss ice crystal size in detail here as it is one parameter impacting the radiative feedback through the longer lifetime of ice crystals in the clouds at high altitudes, which contributes to a stronger positive radiative forcing. We added this to the text.
- c. Changes in the text:

"Combined with the large increase in ICNC from the shift of homogeneous to heterogeneous nucleation, this indicates a large reduction in the size of ice crystals, which would contribute to positive CRE anomalies through longer cloud lifetimes from reduced ice crystal sedimentation."

5. **Comment:** Line 336 - the following paragraph:

This paragraph could be a sub-section with the title , Tropics '.

- a. **Response:** We also agree here and added another sub-section titled "Tropical response"
- 6. **Comment:** Line 340 by up to -10 mg m–3 (Figure 4.4h). (Comment: line break here) Nevertheless, the main effect we find in the tropics is the formation of a large number ...
 - a. **Response:** We agree, and this was implemented in the revised text.

- 7. **Comment:** Line 350 374: This could be a sub-section with the title , Northern Hemisphere '.
 - a. **Response:** We agree. We added a sub-section here in the revised manuscript.
- 8. **Comment:** Line 352: ...we find positive ICNC HET anomalies up to 1000 L⁻¹ at lower levels and a reduction of IWC up to 1.0 mg.m⁻³ (see Figure 4, f,h).
 - a. **Response:** This is incorrect wording. We fixed this in the revised text.

b. Changes in the text:

"Meanwhile, in the NH we find small positive ice crystal radius anomalies in lower levels (p > 600 hPa) for the r0.01 high-seeding case that is consistent with larger ice crystals that sediment more readily. This may be the case in some regions of the NH where we find positive ICNC HET anomalies up to 1000 L⁻¹ at lower levels. This is not reflected in the zonal IWC anomalies at lower levels where we find a reduction of IWC up to 1.0 mg.m⁻³ throughout most of the NH except in the Arctic where it is positive by up to 1.0 mg.m⁻³. However, these low-level IWC signals are insignificant as indicated by the stippling in Fig. 4h."

- 9. **Comment:** Line 359f: This directly influences the large positive LW CRE we find for the r0.01 high- seeding case (Fig. 3 and Tab. 3) Question: maybe better Figure 4 b and Figure 6?
 - a. **Response:** After re-examining the text and the figures, we agree that it is better to reference Figures 4b and 6 in addition to Table 3. This was amended in the revised text.
 - b. Changes in the text:

"This directly influences the large positive LW CRE we find for the r0.01 high- seeding case (Tab. 3, Fig. 4b, and Fig. 6)."

- 10. **Comment:** Line 375 end of Section: This could be a sub-section with the title , Conclusions: global aircraft seeding '.
 - a. **Response:** We agree, but we named the sub-section "Summary of global aircraft seeding".
- 11. **Comment:** Line 394ff: However, we restricted seeding particle emissions further by only seeding during NH wintertime (November-February) as this was suggested to optimize cirrus seeding efficacy (Storelvmo and Herger, 2014; Storelvmo et al., 2014). Comment: Please briefly mention why.
 - a. Response: Good point. We added a short explanation in the text
 - b. Changes in the text:

"However, we restricted seeding particle emissions further by only seeding during NH wintertime (November-February) as this was suggested to optimize cirrus seeding efficacy due to a lack of incident solar radiation during this time period, meaning cirrus exert only a LW warming effect (Storelvmo and Hereger, 2014; Storelvmo et al., 2014)."

12. **Comment:** Line 435: However, as the seeding particles themselves are so small (0.01 μ m), combined with their high number concentration, it is likely that they form numerous ice crystals that remain small due to rapid water vapor consumption such that the average ice crystal size remains roughly the same.

How realistic is the assumption of r = 0.01 μ m (r0.01) and a mass scaling factor of 1000 (high-m given that the consequences for CCT are strong but in nature only INP > ~0.5 um form ice crystals?

 \rightarrow I think this point should be discussed in the paper in some detail.

- a. **Response:** It is a large exaggeration and demonstrates the rather extreme conditions we must simulate in our model in order to achieve significant results. This is included in our discussion section where we describe that our new approach reduces the likelihood of overseeding when using seeding particle sizes similar to previous studies with ECHAM-HAM, but introduces a size bias to achieve a significant signal. We extended the first paragraph in the dicussions section to address this size issue in more detail.
- b. Changes in the text:

"It is unclear whether using such small seeding particles is a feasible approach to represent a real-world seeding scenario as larger particles are typically favored as INPs in the atmosphere. While recent research suggests that smaller particles may be ice active (e.g. pollen and fungal spores, Kanji et al., 2017), it is also unclear if this also applies to cirrus conditions."

13. **Comment:** Line 441: At the same time we find higher rates of heterogeneous nucleation on background dust particles in the stratosphere.

Comment / question: Here and at other places of the manuscript:

Wouldn't it be better to call the region above the mean tropopause 'upper tropopause' instead of stratosphere?

If cirrus clouds form there, then obviously there is enough moisture present but the 'real' stratosphere is dry so that no cirrus clouds can form.

Another possibility is that the tropopause height increases in comparison to the WMO tropopause height due to the induced warming (Fig. 6)? Then, what you called stratosphere could be still upper troposphere? It could be interesting for the reader to discuss that.

- a. **Response:** We did not consider that point. We changed our wording in the revised text when discussing this region to the upper troposphere, lower stratosphere (UTLS). Below we provide how we explain this in the revised text.
- b. Changes in the text:

"As the black line on each plot in Fig. 4 represents the annual mean tropopause height over the five years of our simulations as defined by the World Meteorological Organization (WMO), and to account for tropopause height variations, we refer to this region from now on as the upper-troposphere, lower-stratosphere (UTLS)."

14. **Comment:** Line 473f: Therefore, relative to the unseeded reference case, the new ice crystals forming in the stratosphere in this case are smaller. This behavior also explains the vertical mean ice crystal radius anomalies we found in the global seeding cases in Fig. 5.

It would be good to have a Figure here same as Fig. 4.5, I think seeing the vertical structure if the ice radius anomaly would help understanding the complex processes.

- a. **Response:** This is a good idea, but to not make the main text too repetitive we added a plot for NH wintertime ice crystal anomalies to Appendix C.
- b. Changes in the text;

"Therefore, relative to the unseeded reference case, the new ice crystals forming in the UTLS in this case are smaller. This behavior also explains the vertical mean ice crystal radius anomalies we found in the global seeding cases in Fig. 5. We present a similar vertical ice crystal anomaly plot for our NH wintertime seeding in Appendix C. While the high-seeding case shows the largest reduction in ice crystal radius around 350 hPa it is partially compensated by larger ice particles at lower levels (p > 600 hPa), which is why we find the smallest zonal ice crystal radius anomalies on all levels for this case in Fig. 7b."

- 15. **Comment:** Line 474 end of Subsection: This could be a sub-section with the title , Conclusions: Northern hemisphere-only wintertime seeding '
 - a. **Response:** Similar to your **Comment 10**, we agree but named the sub-section "Summary of northern hemisphere-only wintertime seeding
- 16. **Comment:** Line 476f: Fig. 9 presents the vertical IWC and liquid water content (LWC) anomalies averaged over the NH during Novemver to February for all r0.01 cases with mass emission scaling.

It can be seen from Fig.9 that the positive IWC anomaly is mostly below the mean NH tropopause (~250 hPa), though also above numerous ice particles are injected.

From this one can derive that the ice particles above the WMO tropopause are much smaller so that they do not cause an IWC anomaly, right?

- a. Response: We find that the ice crystals above 250 hPa are smaller, though the peak is around 350 hPa. This is partially related to your Comment 14, where we added an ice crystal anomaly plot to the Appendix. We argue that it is the formation of new ice crystals that are on average smaller than the reference case that leads to the positive IWC anomaly at this level. We find only a small positive ice crystal radius anomaly for the r0.01 high-seeding case at high levels (p < 200 hPa).</p>
- 17. **Comment:** Line 480f: As shown above, this is the result of more numerous and smaller ice crystals that formed on the injected seeding particles. This appears to have an impact on ice crystal sedimentation, ...

Reduced sedimentation because the ice particles are small - I would mention this instead of stating imprecisely 'impact'.

- a. **Response:** We agree, and this was revised to be more precise in the text.
- b. Changes in the text:

"As shown above, this is the result of more numerous and smaller ice crystals that formed on the injected seeding particles. This leads to reduced ice crystal sedimentation, at least for the mid and high-seeding cases, as we find negative IWC anomalies in the lower mixedphase regime."

Comments on Figures/Tables

18. Figure 2: three emissions radii: 0.01 μ m, 0.1 μ m, and 1 μ m' - 0.01 μ m is very small for INP, are they really activated?



- a. Response: We agree that for consistency with the text that the qualitative scaling factors should be included. This is reflected in the new version of the figure in the manuscript. We also find the idea of the 1000 L⁻¹ line is a good idea. This was also added to the new figure. Regarding your comment on size, please see our response to your Comment 2.
- 19. **Figure 3:** Caption: Five-year annual global mean net TOA radiative anomalies (in Wm–2) for the each seeding particle emissions mass scaling factor
 - a. **Response:** Thank you for finding this typo. It was fixed in the revised manuscript.
- 20. **Figure 4**: Please note in the title of the right column that this is the case of high seeding; also, please define the solid and the dashed lines
 - a. **Response:** We agree this should be clearer. Thank you for pointing out that we hadn't defined the lines in the figure caption. This is fixed in the revised manuscript.

21. Figure 5:



- a. **Response:** We agree in line with your other comments on our figure. This was revised in the manuscript.
- 22. Figure 6: Please note in the title of the figure that this is the case of high seeding;
 - a. Response: We agree, and we added this distinction to the title
- 23. Figure 7, Caption: *Comment:* (b) does not show temperature, but Delta_Rice.
 - a. **Response:** Thank you for noting that inconsistency. We fixed this in the revised manuscript.

Figure Comment: please note above or below the figure that the panels are for $r = 0.01 \mu m$

b. **Response:** We agree, and this was added to the revised figure.

24. Figure 8:



- **a.** Response: We disagree with the labels in column a for each ice crystal source as these are included in the titles on the right and explained in the caption. However, the description of these could be clearer. We do agree that the size of the seeding particles could be included as a title. These edits were added to the revised figure.
- b. Changes in the text:

"Five-year northern hemisphere wintertime zonal mean ICNC for in-situ sources of cirrus ice in L⁻¹ for homogeneous nucleation and the sources of heterogeneous nucleation for the unseeded reference case in the first column, and the anomalies for seeding with a factor of 10 (low-seeding, second column), 100 (mid-seeding, third column), and 1000 (high-seeding, fourth column). The first row shows the ICNC from homogeneous nucleation (ICNC HOM). The second, third, and fourth rows each represent the ICNC from heterogeneous nucleation sources for mineral dust (second row, ICNC Dust), soot (third row, ICNC Soot), and seeding particles (fourth row, ICNC Seed). The black line denotes the WMO-defined tropopause and the blue dashed line is the 238 K temperature contour. The stippling denotes insignificant data points at the 95% confidence level according to the independent t-test controlled by the false discovery rate method." 25. Figure 9: the x-axes, aren't they Delta_LWC and Delta_IWC (not LWC, IWC)? Also, please note above or below the figure that the panels are for $r = 0.01 \mu m$.

Caption: Five-year vertical mean anomalies as a function of pressure for (a) IWC and (b) LWC for the NH during the period November to February for seeding with an emissions radius of 0.01 μ m (r0.01) and a mass scaling factor of 1000 (high-m for a mass scaling factor of one (solid line), 10 (dashed line), 100 (dotted line), and 1000 (dot- dashed line). (a) IWC and (b) LWC: The orange dotted line represents the 5-year NH November- February mean temperature vertical profile centred around the homogeneous freezing temperature limit (238 K).

- a. **Response:** Yes, the IWC and LWC should include " Δ " in front. This was changed in the revised manuscript. We disagree with the proposed change in the caption and instead found a new way to incorporate this more clearly.
- b. Changes in the text:

"Five-year vertical mean IWC (a) and LWC (b) anomalies as a function of pressure for the NH during the period November to February for seeding with an emissions radius of 0.01 μ m (r0.01) and a mass scaling factor of 1000 (high-m for a mass scaling factor of one (solid line), 10 (dashed line), 100 (dotted line), and 1000 (dot- dashed line). The orange dotted line represents the 5-year NH November- February mean temperature vertical profile centred around the homogeneous freezing temperature limit (238 K)."

- 26. **Table 1**: You might add to the column 'Freezing method' if all or only a part of the INPs are activated (AF = 1 or AF = f(x); AF: activated fraction; x: Si, T, ...)
 - a. **Response:** We agree with this point. It is defined in the text, but this is a clearer way to display this information. We added an extra row in this column to specify how the number of particles that can become ice active is calculated. We also changed the table caption.
 - b. Changes in the text:

"Adapted from Tully et al. (2022a). A summary of the different aerosol species available for ice nucleation within the in-situ cirrus sub-model. We also present information on the average radius of the particles, the critical ice saturation ratio above which these particles will nucleate ice, the freezing mechanism by which nucleation will occur, and the freezing method within the context of the cirrus scheme following Muench and Lohmann (2020). Under each freezing method we also include the "ice-activity" as a means to define the number of particles in each category that can nucleate ice. Continuous processes are based on the activated fraction (AF) as a function of temperature (T) and S_i, whereas for threshold processes all of the available particles (100%) can nucleate ice unless specified otherwise. "Int. mixed" stands for internally mixed (soluble) aerosol species and "Ext. mixed" stands for externally mixed (insoluble) species. Particle types (i.e. aerosol species) denoted in italics are included as additional processes relative to the base version of our model."

a. **Response:** We believe this comment is for Figure 4. Definitions for the solid and dashed lines were added to the figure captions. For table 3 we added the scaling factor to each line for clarity. Regarding your second comment, it is the cloud fraction and the increase in ICNC of the cirrus that substantially contributes to the TOA anomalies we find, both of which are reflected in Figure 4 as zonal means. In order to keep this table focused, we therefore disagree with adding an additional column for CF. However, we agree this could be added to the text to make it clear that this is a determining factor and to make reference to Figure 4. Please see the response under your **Comment 3**.

Seeding particle emission radius	net TOA	TOA SW	TOA LW	net CRE	SWCRE	LWCRE
μm	No scaling please include the scaling factor					
0.01	0.00 ± 0.91	$\textbf{-0.04} \pm \textbf{0.61}$	0.04 ± 0.34	$\textbf{0.13} \pm \textbf{0.78}$	$\textbf{0.08} \pm \textbf{0.81}$	0.05 ± 0.14
0.1	0.00 ± 0.91	0.01 ± 0.62	$\textbf{-0.01} \pm \textbf{0.34}$	$\textbf{0.00} \pm \textbf{0.78}$	$\textbf{0.02} \pm \textbf{0.81}$	$\textbf{-0.01} \pm \textbf{0.13}$
1	$\textbf{0.02} \pm \textbf{0.91}$	$\textbf{0.03} \pm \textbf{0.61}$	$\textbf{-0.01} \pm \textbf{0.34}$	$\textbf{0.02} \pm \textbf{0.78}$	$\textbf{0.02} \pm \textbf{0.80}$	$\textbf{-0.01} \pm \textbf{0.13}$
	Low-seeding					
0.01	0.31 ± 0.91	$\textbf{-0.37} \pm \textbf{0.61}$	$\textbf{0.68} \pm \textbf{0.34}$	0.60 ± 0.77	$\textbf{-0.18} \pm \textbf{0.81}$	$\textbf{0.78} \pm \textbf{0.14}$
0.1	$\textbf{-0.02} \pm \textbf{0.92}$	$\textbf{-0.01} \pm \textbf{0.61}$	$\textbf{-0.01} \pm \textbf{0.34}$	$\textbf{0.00} \pm \textbf{0.79}$	0.01 ± 0.81	$\textbf{-0.01} \pm \textbf{0.13}$
1	0.00 ± 0.91	0.01 ± 0.61	0.00 ± 0.34	$\textbf{0.00} \pm \textbf{0.79}$	0.01 ± 0.81	$\textbf{-0.01} \pm \textbf{0.14}$
			Mid-seeding			
0.01	$\textbf{2.46} \pm \textbf{0.90}$	-2.34 \pm 0.58	$\textbf{4.80} \pm \textbf{0.36}$	$\textbf{2.57} \pm \textbf{0.77}$	-2.22 \pm 0.76	$\textbf{4.79} \pm \textbf{0.13}$
0.1	$\textbf{-0.05} \pm \textbf{0.90}$	$\textbf{-0.04} \pm \textbf{0.61}$	$\textbf{-0.01} \pm \textbf{0.34}$	$\textbf{0.03} \pm \textbf{0.78}$	0.01 ± 0.81	$\textbf{0.02} \pm \textbf{0.13}$
1	0.00 ± 0.91	0.01 ± 0.61	$\textbf{-0.02} \pm \textbf{0.34}$	$\textbf{0.00} \pm \textbf{0.78}$	0.01 ± 0.80	$\textbf{-0.01} \pm \textbf{0.13}$
			High-seeding			
0.01	$\textbf{5.94} \pm \textbf{0.86}$	-5.05 \pm 0.56	$\textbf{10.99} \pm \textbf{0.36}$	$\textbf{5.04} \pm \textbf{0.72}$	-5.06 \pm 0.75	$\textbf{10.10} \pm \textbf{0.17}$
0.1	$\textbf{-0.02} \pm \textbf{0.90}$	$\textbf{-0.26} \pm \textbf{0.60}$	$\textbf{0.24} \pm \textbf{0.34}$	$\textbf{0.19} \pm \textbf{0.78}$	$\textbf{-0.17} \pm \textbf{0.81}$	$\textbf{0.36} \pm \textbf{0.13}$
1	-0.01 \pm 0.91	0.01 ± 0.62	$\textbf{-0.03} \pm \textbf{0.33}$	$\textbf{-0.01} \pm \textbf{0.78}$	0.01 ± 0.81	$\textbf{-0.02} \pm \textbf{0.13}$
I would suggest to include the IWC / cloud fraction (or what is the driving varialbe?) in the Table, which might give a hint on the reason for the different radiative feedback?						

28. Table 4: see comments on Table 3.

a. Response: Please see response under your Comments 3 & 27.