We sincerely thank the two anonymous reviewers for their thoughtful and constructive comments. In the following, we provide detailed responses to each comment and indicate how they have been addressed in the revised manuscript. The reviewers' comments are shown in blue, our replies are in black, and the corresponding revisions in the manuscript are highlighted in red (line numbers refer to the tracked version of the revised manuscript).

Recommendation: Return to Authors for Major Revisions

Overview

This manuscript has improved compared to previous versions—thank you for your continued efforts. However, the presentation still lacks clarity and logical flow, particularly in key technical sections. Below, I provide a set of major and minor comments that should be addressed to improve the overall quality and readability of the paper.

Major comments/questions:

1. Clarification and Completeness of the K22 Scheme Description

The presentation of the K22 scheme still requires significant clarification. Several key conceptual and technical aspects remain unclear:

• Scope of K22: The K22 scheme is designed for cirrus clouds. To my understanding, it does not apply to freezing processes in mixed-phase clouds. Please clarify this explicitly.

Reply: Thank you very much for your helpful and constructive comments. Yes, the K22 scheme is designed only for cirrus clouds and does not apply to freezing processes in mixed-phase clouds.

In CAM6, cirrus clouds are defined as the clouds with temperatures below -37 °C and mixed-phase clouds are defined as the clouds with temperatures between 0 and -37 °C. The freezing processes in cirrus clouds and mixed-phase clouds are treated differently. Heterogeneous ice nucleation in mixed-phase clouds is based on the classical nucleation theory including immersion, deposition and contact freezing with rates depending on the properties of mineral dust and black carbon aerosols (Hoose et al., 2010; Wang et al., 2014).

The relevant paragraph has been modified as follows (line 92-96):

"In CAM6, cirrus clouds are defined as the clouds with temperatures below -37 °C and mixed-phase clouds are defined as the clouds with temperatures between 0 and -37 °C. Ice nucleation in cirrus clouds is treated differently (see section 2.2) from that in mixed-phase clouds. Ice nucleation in mixed-phase clouds is treated based on the classical nucleation theory including immersion, deposition and contact freezing with rates

depending on the properties of mineral dust and black carbon aerosols (Hoose et al., 2010; Wang et al., 2014). "

• Equations (1) – (5): These equations appear to describe heterogeneous ice nucleation only. If so, why is homogeneous freezing not included in this section? Please incorporate the corresponding parameterization for homogeneous nucleation. Furthermore, how is INP activation efficiency for homogeneous freezing quantified?

Reply: Following the reviewer's comment, we have provided an explanation of homogeneous nucleation in the revised manuscript. Please note that homogeneous nucleation refers to the spontaneous freezing of aerosol (sulfate) solution droplets, and does not involve INPs (e.g., dust, black carbon). The relevant paragraph has been modified as follows (line 100-115):

"In the K22 parameterization, the number of activated solution droplets (n_{homo}) over time is calculated based on the freezing rate (j), following the expression:

$$n_{homo} = n_{sulfate}[1 - exp(\int -jdt)] \tag{1}$$

 $n_{sulfate}$ is the initial number concentration of sulfate solution droplets, the freezing rate j is determined using the liquid water volume (V) of the solution droplet population and a rate coefficient (J) derived from a water activity-based formula (Koop et al., 2000) (j=VJ). The parameterization assumes a monodisperse size distribution of solution droplets with radius of 0.25 µm, neglecting the presence of a small amount of soluble material in the droplets. Vertical velocity (w), supersaturation with respect to ice (S_i), and temperature (T) significantly influence the water activity so that $J=J(w,S_i,T)$ (Baumgartner et al., 2022; Kärcher et al., 2022; Liu & Penner, 2005). The thermodynamic threshold S_{hom} for homogeneous freezing to take place is estimated through an iterative process in which the deposition growth of ice crystals from previously frozen solution droplets reduces the supersaturation. This quenching process is a function of T, w, and the mean droplet size (Kärcher et al., 2022). Once S_{hom} is determined, the number concentration of newly nucleated ice crystals is computed using S_{hom} , S_i and effective updraft speed (see Equation (6) below). More detailed information can be found in Kärcher et al. (2022)."

 Supersaturation and Equation (2): Equation (2) relies on supersaturation (S) to compute Φ. However, S is determined from Equation (3), which seems to be highly nonlinear in S. What numerical method or approximation is used to solve Equation (3)?

Reply: The ice supersaturation threshold at heterogeneous activation-relaxation is determined by numerical iteration when the dS/dt=0 (i.e., the production and loss of supersaturation in equation (3) (now equation (4)) are equal) and used to compute the Φ from INPs. If homogeneous nucleation also occurs, the ice supersaturation threshold at

homogeneous activation-relaxation determined similarly is used to compute the Φ from INPs.

We added a sentence (line 155-158):

"The ice supersaturation threshold at heterogeneous activation-relaxation is determined by numerical iteration when the dS/dt=0 (i.e., the production and loss of supersaturation in equation (4) are equal) and used to compute the Φ from INPs in equation (3). If homogeneous nucleation also occurs, the ice supersaturation threshold at homogeneous activation-relaxation determined similarly is used to compute the Φ from INPs."

• Role of Vertical Velocity: The treatment of vertical velocity is unclear. My understanding is that an effective vertical velocity weff is derived from Equation (5) and passed into Equation (3), with a steady-state assumption dSdt=0. If so, how are w(q,het) and w(q,pre) derived? Is Equation (4) used in this context? Please specify how the the right-hand side of Equation (4) are quantified.

Reply: Yes, weff derived from equation (5) (now equation (6)) is used to calculate ice number from homogeneous nucleation. To calculate w(q,het), the loss term due to the deposition of water vapor onto ice crystals formed from heterogeneous nucleation, denoted as $L_{q,het}$, must first to be determined:

$$L_{q,het} = \sum_{k=1}^{K} \frac{n_k}{n_{sat}} \frac{dN_k}{dt}$$

where the index k denotes an INP class, with associated ice number concentrations n_k resulting from nucleation of the fraction of INPs that become ice-active within a supersaturation interval ΔS_k . N_k is the number concentration of water molecules per ice crystal formed from INPs in each supersaturation class. The water molecule number concentration at ice saturation n_{sat} is obtained from Murphy and Koop (2005). The rate of change in the number of water molecules per ice crystal is given by $\frac{dN_k}{dt} = 4\pi r_k D_k n_{\text{sat}} S$, where r_k is ice crystal radii, assuming a spherical volume centered on the INP core: $r_k = (r_c^3 + \frac{vN_k}{4\pi/3})^{1/3}$. Here, v is the volume of a single water molecule in ice and r_c is the radius of the dry aerosol particle core (assumed to be 0.2 µm). The effective diffusivity D_k is given by: $D_k = D_v(\frac{r_k}{r_k+l} + \frac{d}{\alpha_k r_k})^{-1}$, where D_v is the water diffusion coefficient in air, l is the jump distance for water molecules (approximately equal to the mean free path), $d=4D_v/v$ is the diffusion length scale, with v being the mean thermal speed of water, and α_k is the deposition coefficient specific to the ice crystals formed in the supersaturation interval ΔS_k .

The quenching velocity due to heterogeneous nucleation w(q,het) is then calculated as:

$$w(q, het) = \frac{L_{q,het}}{a(S+1)}$$

To compute the quenching velocity due to pre-existing ice, w(q,pre), the loss term due to the removal of water vapor onto pre-existing ice crystals $L_{q,pre}$ is calculate as:

$$L_{q,pre} = \int_0^s \frac{4\pi}{v n_{sat}} \frac{dn}{ds'} \left(\int_{\tau(s')}^{t(s)} r^2 \frac{dr}{dt} dt \right) ds'$$

Finally, w(q,pre) is:

$$w(q, pre) = \frac{L_{q,pre}}{a(S+1)}$$

The relevant paragraph has been modified as follows (line 137-158):

"When $\frac{ds}{dt} = 0$ in Equation (4), we can define the quenching velocity $w_{q,pre}$ due to pre-existing ice crystals as:

$$w_{q,pre} = \frac{\int_{0}^{s} \frac{4\pi}{v n_{sat} ds'} (\int_{\tau(s')}^{t(s)} r^{2} \frac{dr}{dt} dt) ds'}{a(s+1)},$$
 (5)

where the loss term of water vapor includes the contribution from pre-existing ice. The quenching velocity due to heterogeneous ice nucleation $w_{q,het}$ can be calculated similarly based on Kärcher et al. (2022), using the equation: $w_{q,het} = \frac{L_{q,het}}{q(s+1)}$. Here, $L_{q,het}$ is the loss term due to the deposition of water vapor onto ice crystals formed from heterogeneous nucleation: $L_{q,het} = \sum_{k=1}^{K} \frac{n_k}{n_{sat}} \frac{dN_k}{dt}$. The index k denotes an INP class, with corresponding ice number concentrations n_k that result from nucleation of the fraction of INPs that become ice-active within a supersaturation interval ΔS_k . N_k represents the number concentration of water molecules per ice crystal formed from INPs in each supersaturation class. The water molecule number concentration at ice saturation n_{sat} is obtained from Murphy and Koop (2005). The rate of change in the number of water molecules per ice crystal is given by $\frac{dN_k}{dt} = 4\pi r_k D_k n_{sat} S$, where r_k is ice crystal radii, assuming a spherical volume centered on the INP core: $r_k = (r_c^3 + \frac{vN_k}{4\pi/3})^{1/3}$. In this expression, v is the volume of a single water molecule in ice, and r_c is the radius of the dry aerosol core (assumed to be 0.2 μ m). The effective diffusivity D_k is given by: $D_k =$ $D_v(\frac{r_k}{r_k+l}+\frac{d}{\alpha_k r_k})^{-1}$, where D_v is the water diffusion coefficient in air, l is the jump distance for water molecules (approximately equal to the mean free path), $d = 4D_v/v$ is the diffusion length scale, v is the mean thermal speed of water molecules, and α_k is the deposition coefficient specific to ice crystals formed within the supersaturation interval $\Delta S_{\rm k}$."

• Homogeneous Freezing – Liquid Water Volume (V): Line 94 refers to a required liquid water volume V. How is this calculated? Is it based on an assumed droplet

size (e.g., $0.25~\mu m$ as mentioned in Line 97) and estimated droplet number concentration? Please describe the approach in detail.

Reply: The droplet population is assumed to be a lognormal droplet distribution with modal radius of 0.25 μ m, and geometric standard deviation of 1 (i.e., monodisperse). V is the volume of a solution droplet. The droplet number concentration is assumed to be 500 per cubic centimeter. The estimated activated droplet number concentration is calculated by $n_{homo} = n_{sulfate}[1 - exp(\int -jdt)]$.

The relevant paragraph has been modified as follows (line 105-107):

"The parameterization scheme assumes a monodisperse size distribution of solution droplets with radius of 0.25 μ m, neglecting the presence of a small amount of soluble material in the droplets."

Please address each of the above points clearly and systematically. These questions are fundamental for assessing whether the authors have a solid understanding of the K22 scheme and its implementation. Additional technical details can be placed in the Supporting Information if necessary.

2. Analysis of Freezing Frequencies and Vertical Velocity Contributions

It would significantly strengthen the manuscript if the authors analyzed the relative frequencies of homogeneous versus heterogeneous freezing events predicted by the K22 scheme. Specifically:

- Show the distribution of weff (from Eq. 5) as a function of latitude and altitude.
- Examine the role of orographic gravity waves (OGW) in producing positive weff values.
- Clarify whether w(q,het) is calculated as w(q,het)=w-w(q,pre). If so, provide the frequency and latitudinal/vertical distribution of w(q,het).

This analysis will offer deeper insight into the competition between homogeneous and heterogeneous freezing. Currently, the paper only discusses changes in ice crystal number concentration (Ni), but the vertical velocity distribution—being the root cause of these changes—deserves direct analysis.

Reply: Thank you very much for your helpful and constructive comments. It is a good idea to check homogeneous and heterogeneous freezing frequencies in the K22 scheme based on the effective vertical velocity w_{eff} and quenching speed due to heterogeneous nucleation.

• The distribution of w_{eff} (from Eq. 6, formerly Eq. 5) in the K22_OGW-Climo experiment as a function of latitude and altitude is shown in Fig. R1-1. The figure is included in the supplemental figures. The relevant paragraph has been modified as follows (line 302-304):

"In the K22_OGW-Climo experiment, strong w_{eff} is found over mid- and high latitudes (Fig. S1), with the large positive w_{eff} occurring primarily over the high mountain regions (Fig. S2). This pattern indicates the important contribution of OGWs in producing positive w_{eff} values."

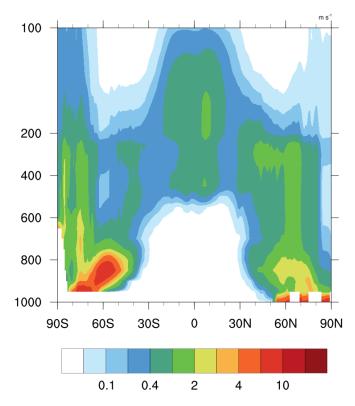


Figure R1-1. Annual mean w_{eff} (unit: m s⁻¹) as a function of latitude and altitude from the K22-OGW-Climo experiment.

• Figure R1-1 shows strong w_{eff} over mid- and high latitudes. In addition, w_{eff} at 250 hPa is presented in Fig. R1-2. The large positive w_{eff} over mid- and high latitudes is primarily located over the high mountains, indicating the contributing role of OGWs in producing positive w_{eff} values. The w_{eff} spatial distribution indicates the frequency of homogeneous nucleation occurrences.

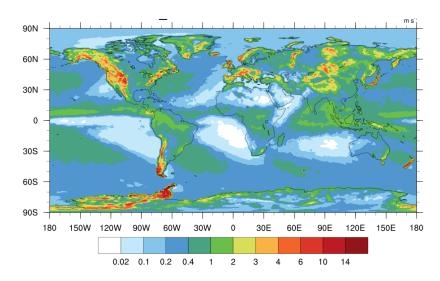


Figure R1-2. Annual mean w_{eff} at 250 hPa from the K22-OGW-Climo experiment (unit: m s⁻¹).

• The quenching speed w(q,het) is not equal to w - w(q,pre) in most cases because the effective updraft speed is defined as $w_{eff}=w - w(q,het) - w(q,pre)$. The zonal mean w(q,het) is shown in Fig. R1-3, which indicates that strong quenching effect due to heterogeneous ice nucleation primarily occurs in the lower troposphere, especially in the Northern Hemisphere and in the mid-latitudes of the Southern Hemisphere. High concentrations of coarse mode dust are found in the lower troposphere, especially in mid-latitudes (Fig. R1-4). Fig. R1-5 shows w(q,het) and coarse mode dust number concentrations at 350 hPa. A pronounced concentration coarse mode dust is found over Tibetan Plateau, corresponding one high value region of w(q,het). This suggests that elevated coarse mode dust number concentrations are necessary for the occurrence of strong w(q,het).

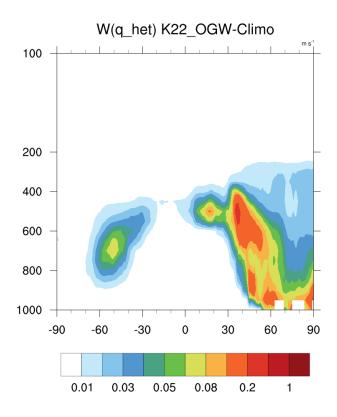


Figure R1-3. Zonal mean $w(q_het)$ in the K22-OGW-Climo experiment (Unit: m s⁻¹).

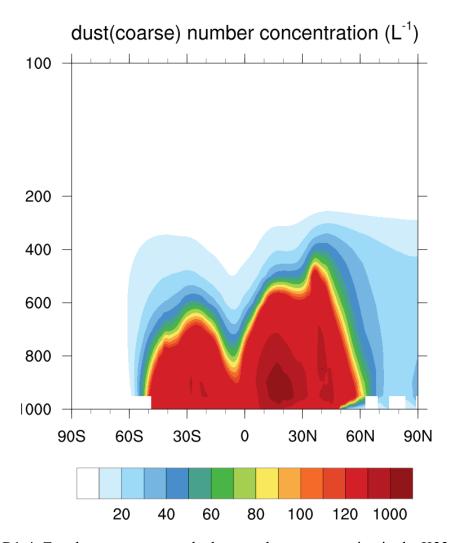


Figure R1-4. Zonal mean coarse mode dust number concentration in the K22-OGW-Climo experiment (Unit: L⁻¹).

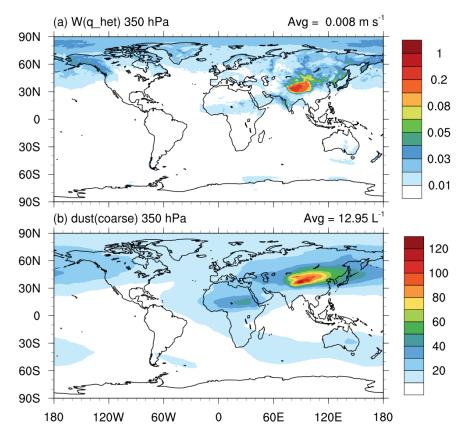


Figure R1-5. $w(q_het)$ and coarse mode dust at 350 hPa in the K22-OGW-Climo experiment.

Minor comments/questions:

1. Lines 1–2: If the study explores all ice formation pathways, sources like detrainment should be mentioned. The title currently suggests the focus is limited to INP activation. Consider emphasizing "Ice Crystal Formation from INP Activation in Cirrus Clouds" for clarity.

Thank you very much for your constructive comment. Since this study focuses on ice nucleation in cirrus clouds by using two ice nucleation schemes, we would like to keep the current title. However, we modified the "ice nucleation" to "ice crystal formation" for the consideration of all ice formation pathways in the first sentence in the paragraph as follows (line 7-8):

"Ice crystal formation in cirrus clouds is poorly understood, and its representation remains a challenge in global climate models."

2. Lines 18–20: The sentence beginning with "However..." is unclear and should be rewritten for better readability.

Thank you very much for your comment. The relevant sentence has been modified as follows (line 19-21):

- "Due to its distinct competition parameterizations, the K22 scheme exhibits less contribution from minor ice sources (convection detrained and turbulence-induced)."
- 3. Lines 20–22: The impact summary is too weak. This study could help clarify regional and dynamical controls on INP activation. Please strengthen the impact statement.

Thank you very much for your constructive comment. The relevant paragraph has been modified as follows (line 21-24):

- "This underscores the significance of competition mechanisms within ice nucleation schemes and helps clarify regional and dynamical controls on ice sources in cirrus clouds."
- 4. Lines 25–26: The claim that cirrus clouds "warm the planet" needs more explanation. While cirrus do absorb longwave radiation, the extent to which they warm the surface depends on their net radiative forcing. Please clarify.

Thank you very much for your helpful and constructive comment. The relevant paragraph has been modified as follows (line 27-28):

- "They can also absorb terrestrial longwave radiation, thereby contributing to warming the atmosphere."
- 5. Lines 26–27: The phrase "cirrus clouds determine cloud radiative forcing" is too strong. Other cloud types (e.g., stratocumulus) also contribute significantly. Please moderate the statement.

Thank you very much for your helpful and constructive comment. The relevant paragraph has been modified as follows (line 28-30):

- "The balance between these two opposite processes is greatly influenced by the microphysical properties of ice crystals in cirrus clouds, which in turn affects the net cloud radiative forcing."
- 6. Lines 58–62: Please clarify the distinction between uncertainty in supersaturation and INP activation efficiency. In my view, supersaturation is an external factor affecting INP efficiency (which also depends on chemical composition). The aerosol number concentration determines the number of INPs. What exactly is meant by "activation efficiency" in this context as an independent factor for INP activation?

Thank you very much for your helpful and constructive comment. By the activation efficiency we meant activation fraction at a given condition, which depends on physical and chemical properties (e.g., morphology, chemical composition) of INPs. The relevant sentence has been modified as follows (line 60-63):

"Limited knowledge of the number concentration and properties (e.g., morphology, chemical composition) of INPs in the upper troposphere complicates the model prediction of cirrus clouds microphysical properties (Kärcher et al., 2022; Knopf & Alpert, 2023)."

7. Lines 68–70: Does the prior sentence imply that the K22 scheme is already implemented in CAM6? Please confirm.

Thank you very much for your helpful and constructive comment. Sorry that it was not clear. The K22 scheme is implemented in CAM6 for the first time in this study. The relevant sentence has been modified as follows (line 68-71):

"In this study, a new parameterization (Kärcher, 2022), referred to as K22, that encompasses homogeneous nucleation, heterogeneous nucleation, their interactions, and competition with preexisting cirrus ice, is integrated into CAM6. We further evaluate its effects on ..."

8. Lines 85–88: I use CESM as well but am not familiar with INP-MAM4 interactions as described. Could you specify the CESM code version and point to the relevant code section? Also, conceptually, converting INPs to cloud borne aerosols seems problematic, as cloud-borne aerosols typically refer to CCN, not IN. I may be mistaken, but showing the code would clarify this.

Thank you very much for your helpful and constructive comment. In CESM2, cloud borne aerosols have been extended from CCN in warm clouds to including INPs for ice nucleation in cold clouds. Upon ice nucleation, INPs will be converted to cloud-borne aerosols as well. Future model development could separate "cloud borne aerosols" into "droplet-borne aerosols" for warm clouds and "ice-borne aerosols" for cold clouds.

The version of CESM used in this study is CESM2.2.0. The relevant code section can be found in components/cam/src/physics/cam/nucleate_ice_cam.F90.The source code means that the INPs are converted to cloud borne once they are nucleated.

```
! Move aerosol used for nucleation from interstial to cloudborne,
! otherwise the same coarse mode aerosols will be available again
! in the next timestep and will supress homogeneous freezing.
if (prog_modal_aero .and. use_preexisting_ice) then
if (separate_dust) then
call endrun('nucleate_ice_cam: use_preexisting_ice is not supported in separate_dust mode (MAM7)')
endif
ptend%q(i,k,cnum_idx) = -(odst_num * icldm(i,k))/rho(i,k)/1e-6_r8/dtime
cld_num_coarse(i,k) = cld_num_coarse(i,k) + (odst_num * icldm(i,k))/rho(i,k)/1e-6_r8
```

```
ptend\%q(i,k,cdst\_idx) = - \ odst\_num \ / \ dst\_num \ * \ icldm(i,k) \ * \ coarse\_dust(i,k) \ / \ dtime cld\_coarse\_dust(i,k) = cld\_coarse\_dust(i,k) + odst\_num \ / \ dst\_num \ * icldm(i,k) \ * \ coarse\_dust(i,k) end \ if "
```

9. Lines 97–98: Please include the homogeneous freezing parameterization in the paper. Especially, how is the liquid water content (V) calculated, and what assumptions are made (e.g., droplet size = $0.25 \mu m$)? How is droplet number determined? This will help readers follow without referring back to Karcher et al. (2022).

Please see our response to your previous major comment above. V is the volume of a solution droplet. The droplet number concentration is assumed to be 500 per cubic centimeter. The estimated activated droplet number concentration is calculated by $n_{homo} = n_{sulfate}[1 - exp(\int -jdt)]$.

10. Lines 114–115: How exactly is water vapor removed? Is it due to deposition on preexisting ice, newly formed ice, or other processes like entrainment/detrainment? Please clarify and list the quantification of all the terms.

Thank you very much for your helpful and constructive comment. The water vapor is removed by deposition onto newly nucleated ice or onto pre-existing ice (i.e., ice formed from previous time steps). The relevant paragraph has been modified as follows (line 130-131):

"The removal of water vapor can be caused by the deposition onto newly nucleated ice crystals or onto pre-existing ice crystals."

11. Lines 150–152: I still find the competition between homogeneous, heterogeneous, and pre-existing ice formation difficult to follow. Is this primarily reflected in Equation (5)? If weff<0, does that imply no homogeneous freezing?

Thank you very much for your helpful and constructive comment. The K22 scheme represents the competition between homogeneous, heterogeneous and pre-existing ice by quenching velocities, reflected in Eq. (5) (now Eq. (6)). We have added the equations for calculating the quenching velocities from heterogeneous nucleation and from pre-existing ice in the revised manuscript. Please see our response to your major comment #1 above.

Yes. If $w_{\text{eff}} < 0$, this means that no homogeneous freezing happens.

12. Lines 179–181: Were these simulations free-running or nudged? If they were free-running, I'm concerned that large-scale meteorological differences may limit meaningful comparison with field campaign observations.

Thank you very much for your comment. The 6-year climatology simulations are freerunning, while the simulations related to two flight campaigns are wind (UV)-nudged towards the MERRA2 reanalysis. We added a sentence in the revised manuscript (line 221-222):

"For the nudged simulations for the two field campaigns (Table 2), the modelled horizontal winds are nudged towards the MERRA2 reanalysis data."

Reference:

- Baumgartner, M., Rolf, C., Grooß, J. U., Schneider, J., Schorr, T., Möhler, O., Spichtinger, P., & Krämer, M. (2022). New investigations on homogeneous ice nucleation: the effects of water activity and water saturation formulations. *Atmos. Chem. Phys.*, 22(1), 65-91. https://doi.org/10.5194/acp-22-65-2022
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- Murphy, D. M., & Koop, T. (2005). Review of the vapour pressures of ice and supercooled water for atmospheric applications. *Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography*, *131*(608), 1539-1565, ISSN = 0035-9009, DOI = https://doi.org/1510.1256/qj.1504.1594.

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Thanks to the authors for addressing the reviewer comments. I recommend accepting the manuscript after the following minor issues are resolved.

1. L19: The phrase "less contribution" seems to better fit the context than "less competition". The authors may consider rewording it.

Thank you very much for your suggestion. We reworded "less competition" to "less contribution". The relevant paragraph has been modified as follows (line 19-21):

"Due to its distinct competition parameterizations, the K22 scheme exhibits less contribution from minor ice sources (convection detrained and turbulence-induced)."

2. L391-392: Should the positions of "SWCF" and "LWCF" be swapped? Figures R2-1 (d) and (e) show that the average LWCF and SWCF are -0.506 and 2.950 W/m2, respectively. Please check.

Thank you very much for your comment. We have carefully re-checked SWCF and LWCF. The SWCF and LWCF values from both the K22 and LP05 schemes are presented in Figure R2-1. The distributions of SWCF and LWCF appear similar between both schemes. Interestingly, the differences in SWCF and LWCF changes between the two schemes contradict the conventional expectation that more frequent cirrus clouds in K22 compared to LP05 should result in a stronger positive LWCF and a more negative SWCF. These unexpected results, derived from the RRTMG radiation scheme, are likely influenced by the presence of thin cirrus clouds and the high solar zenith angles typical of mid- and high-latitude regions. The cirrus clouds in question are sufficiently thin to allow longwave radiation to pass through. Additionally, the high solar zenith angles may enhance the scattering of shortwave radiation within ice crystals, allowing more shortwave radiation to reach the surface instead of being reflected back to space. Moreover, the surface reflectivity may also play an important role. At high solar zenith angles, ocean surfaces can reflect 50-80% of incoming solar radiation, and snow-covered land can reflect 80-90% sunlight, while cirrus clouds can only reflect 10-40%. These combined factors may explain the unexpected results of positive change of SWCF and negative change in LWCF from LP05 to K22 schemes.

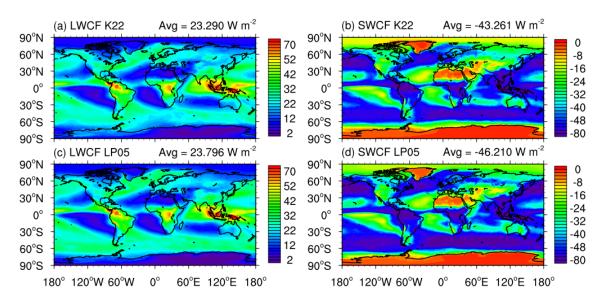


Figure R2-1. The SWCF and LWCF in both the K22 (upper panel) and LP05 schemes (lower panel).