

Response to Anonymous Referee #1

egusphere-2025-5850: “Global Modeling of Ice Nucleating Particles of Multiple Aerosol Species and Associated Cloud Radiative Effects” by K. Kawai, Z. Ren, and H. Matsui

Thank you very much for carefully reading our manuscript and providing valuable comments. We have revised the manuscript by taking your comments into account. Below, we describe our point-by-point responses to your comments. A revised manuscript with tracked changes has been uploaded.

Referee’s comment 1-1:

Section 1: There is a recent study by Imura and Suzuki (doi:10.1175/JCLI-D-24-0335.1) that investigated climatic impact of differing INP-temperature spectra through precipitation process, which I think is relevant to the scope of this study. The authors should discuss the paper in the context described in introduction.

Response:

As suggested, we have added Imura and Suzuki (2025) to Section 1 (Introduction) as follows (Lines 30–32): “*In addition, differences in the temperature-dependent ice nucleating properties of INPs can lead to substantial variations in cloud microphysical processes and their associated climatic impacts (Imura and Suzuki, 2025).*”

Imura, Y., and Suzuki, K.: Evaluation of Climatic Impacts of Ice-Nucleating Particles through Precipitation Process with a GCM, J. Climate, 38, 2659–2677, <https://doi.org/10.1175/JCLI-D-24-0335.1>, 2025.

Referee’s comment 1-2:

Section 2.1: It would be useful to add the plot that compares how INP number concentration depends on temperature, or the so-called INP-temperature spectra for various aerosol species treated in this study. Such a plot would be very nice to illustrate how different

aerosol species newly considered in this study have different efficiencies as INPs. The plot should also include the observationally constrained function of bacteria described in Section 2.3.

Response:

As suggested, we have added plots of the ice nucleating abilities of the aerosol species considered as INPs in this study and moved Fig. S1 to Fig. 1 (revised version) as follows:

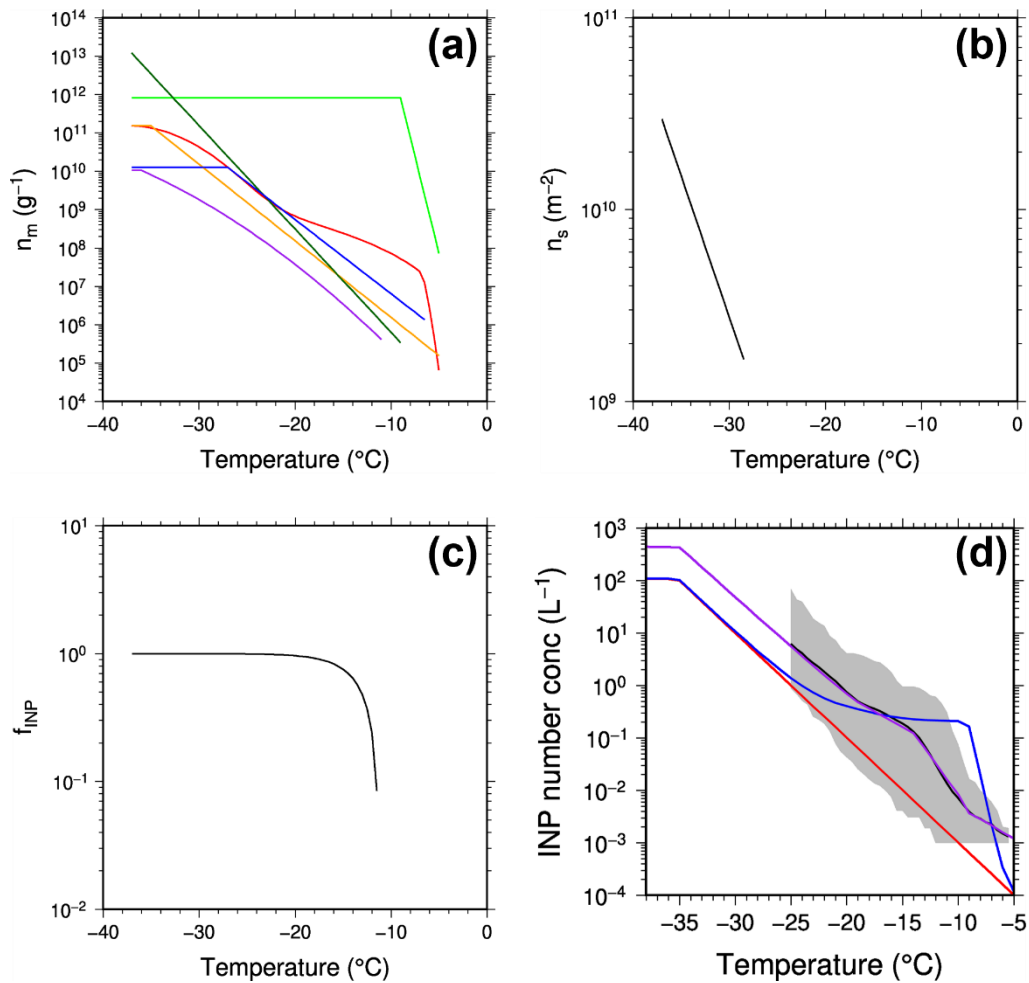


Figure 1. Ice nucleating abilities of aerosol species and types acting as INPs in this study. (a) The red, orange, light green, green, blue, and purple lines represent the ice nucleation active site density per unit mass (n_m) of Arctic dust, non-Arctic dust, bacteria, pollen, MOA, and biomass burning BC, respectively. The n_m of non-Arctic dust and MOA were derived using the size distributions of their emitted particles assumed in our model. (b) The ice nucleation active site density per unit surface area (n_s) of *Cladosporium sp.* fungal spores. (c) The

activated fraction of Mortierella alpina fungal spores. (d) Annual mean INP number concentrations as a function of freezing temperature observed at Tokyo Skytree, Japan, from August 2016 to July 2017 (Tobo et al., 2020) (black) and simulated for the corresponding location and period for dust only (red) and for all INP sources in the Base simulation (blue) and the observationally constrained simulation (purple). The gray shading indicates the range of observed INP number concentrations.

Referee's comment 1-3:

Line 91-93: I don't understand what this sentence means. Why do you talk about the treatment of bioaerosols, MOA and BC here in this subsection for dust? Please clarify what "the same computational approach" means.

Response:

For dust, bioaerosols, MOA, and BC, using the ice nucleating abilities of each species, the INP number concentrations of each species are calculated online as a function of air temperature and the mass or number concentrations of each species in the presence of liquid water droplets. We have moved this explanation from Section 2.2.1 (Dust) to Section 2.1 (Global climate-aerosol model) as follows (Lines 85–90): "*Using the ice nucleating abilities of each species, the INP number concentrations of each species are calculated online as a function of air temperature (between -37°C and -5°C) and the mass or number concentrations of each species (both interstitial-phase and cloud-phase) in the presence of liquid water droplets. In this study, we report the number concentrations of INPs within clouds, which we equate to the grid-mean INP number concentrations multiplied by the fractions of stratus clouds. This calculation is consistent with the CAM5 cloud microphysics scheme (Gettelman et al., 2010; Morrison and Gettelman, 2008).*"

Referee's comment 1-4:

Line 104-106: Is there no temperature dependence for bioaerosols assumed here?

Response:

Thank you for your comment. In our treatment of bioaerosols, only a prescribed fraction of bioaerosol particles is assumed to be capable of acting as INPs (4%, 100%, 29%, or 8%), and this fraction itself is not temperature dependent. For these bioaerosol particles, the temperature dependence of n_m or n_s (the ice nucleation active site density per unit mass or surface area, respectively) is explicitly calculated (Diehl and Mitra, 2015; Hummel et al., 2018). We have added this point to the text as follows (Lines 148–150): “*The temperature-dependent ice nucleating abilities of 4 % of bacteria (Pseudomonas syringae) and 100 % of pollen (tree pollen) are based on Diehl and Mitra (2015) (Fig. 1a). We assumed that 29 % of Cladosporium sp. fungal spores and 8 % of Mortierella alpina fungal spores act as INPs with temperature dependence following Hummel et al. (2018) (Fig. 1b and 1c).*”

Referee’s comment 1-5:

Line 134-136: Do these multiplication factors mean the factors relative to the Base simulation?

Response:

Yes, these multiplication factors mean the factors relative to the ice nucleating ability of bacteria used in the Base simulation. We have clarified this point as follows (Lines 182–184): “*The ice nucleating ability of bacteria used in the Base simulation was multiplied by a scaling factor of $10^{3.2}$ at ≤ -35 °C, $10^{0.2}$ at -19 °C, $10^{-0.3}$ at -14 °C, $10^{-1.8}$ at -9 °C, and $10^{1.8}$ at -5 °C, with a lognormal interpolation between these temperatures.*”

Referee’s comment 1-6:

Line 145: “between all-sky and clear-sky conditions”: Is this not a “clean-sky” CRE? That is, does the cloud radiative forcing here includes the effect of light-absorbing aerosols on cloud radiative effect? Please clarify.

Response:

CRE is calculated under aerosol-containing atmospheric conditions (not “clean-sky”). However, because aerosol fields are nearly identical between two simulations (e.g., dust INPs

×10 simulation and the Base simulation), most aerosol radiative effects may cancel out.

Referee's comment 1-7:

Line 290-292: I don't understand what this sentence means. Can you explain more clearly how the fewer total INPs, more water droplets and fewer ice crystals at the temperature warmer than -10°C relate to the higher sensitivity of bioaerosols than dust?

Response:

We want to show in this sentence that mixed-phase clouds typically contain relatively few ice crystals and a large amount of supercooled water droplets at temperatures warmer than approximately -10°C where bioaerosols dominate INP activity. Under such conditions, even a small increase in INP number concentration can efficiently enhance ice formation, which in turn promotes the conversion of water droplets to ice crystals via the Wegener–Bergeron–Findeisen process. As a result, bioaerosols could exert a relatively large impact on cloud microphysical properties and radiative effects despite their smaller INP number concentrations compared to dust. We have revised the sentence to clarify this interpretation as follows (Lines 346–351): *“This difference in sensitivity might be related to the fact that, at temperatures where bioaerosols dominate INP activity (warmer than approximately -10°C), mixed-phase clouds typically contain relatively few ice crystals and abundant supercooled water droplets. Under such conditions, an increase in INPs could efficiently enhance ice formation and promote the conversion of water droplets to ice crystals, leading to a stronger impact on cloud properties.”*

Referee's comment 1-8:

Line 294-295: Does this mean that the dust and bioaerosol contributions to overall CRE are mostly linear?

Response:

Thank you for your comment. This sentence means that the global-mean CRE in the Base simulation can be largely explained by the sum of the dust and bioaerosol contributions. However, we avoid using the term “linear” here because it could be interpreted as implying that

the cloud response is proportional to changes of INP number concentrations.

Referee's comment 1-9:

Line 301-303 and Table 2: The table shows only the “All INP sources” value of CRE. Are the CREs from aerosol species other than bacteria are same between the observationally constrained and Base simulations? Namely, does the difference in CRE from the all INP sources between the two simulations (0.64 vs 0.24 Wm⁻²) come from only the bioaerosol-induced CRE? Please clarify.

Response:

The CREs of all aerosol species other than bacteria via INPs are almost the same between the Base and observationally constrained simulations. Although the differences in CREs between the Base and observationally constrained simulations result from the treatment of bioaerosols, this constraint should be interpreted as reflecting uncertainties in multiple factors, including not only the emission and ice nucleating ability of bacteria, but also INPs from other aerosol species and unknown sources. We explain this point in Section 3.4 as follows (Lines 364–366): “*Although we constrain the ice nucleating ability of bacteria to the INP observations in Tokyo, the differences in CREs between the Base and observationally constrained simulations may reflect uncertainties in various factors, including not only the emission and ice nucleating ability of bacteria, but also INPs from other aerosol species and unknown sources.*”

Referee's comment 1-10:

Figure 3 caption: orange -> yellow. To my eye, the color of dust looks more like yellow rather than orange.

Response:

As suggested, we have changed “orange” to “yellow” in the caption of Figure 5 (revised version) as follows (Lines 277–279): “*Figure 5. Fractions of dust (yellow), bioaerosols (green), MOA (blue), and BC (gray) in the annual and global mean vertically integrated total INP number concentrations (shown on the right) in (a) the whole atmosphere (2–1000 hPa) and (b)*

the upper (100–400 hPa), middle (400–700 hPa), and lower troposphere (700–1000 hPa).”

Response to Anonymous Referee #2

egusphere-2025-5850: “Global Modeling of Ice Nucleating Particles of Multiple Aerosol Species and Associated Cloud Radiative Effects” by K. Kawai, Z. Ren, and H. Matsui

Thank you very much for carefully reading our manuscript and providing valuable comments. We have revised the manuscript by taking your comments into account. Below, we describe our point-by-point responses to your comments. A revised manuscript with tracked changes has been uploaded.

Referee’s comment 2-1:

Lines 21 – 26: Are there studies showing how increasing the ice fraction decreases cloud lifetime and cloud fraction due to the higher fall speeds of ice particles? This will affect the CRE. If so, this process with references should be mentioned. Relevant references might be Mitchell et al. (2008, GRL) and Eidhammer et al. (2017, J. Climate, p. 618).

Response:

Thank you for suggesting these references. An increase in the ice fraction in mixed-phase clouds can lead to a reduction in cloud lifetime and cloud fraction because ice crystals generally have larger sizes and higher falling speeds than water droplets. These characteristics of ice crystals enhance formation of precipitation and the removal of condensate. This process further influences cloud radiative effects. We have added this explanation with the suggested references to the Introduction as follows (Lines 27–30): “*This shift toward the ice phase can decrease cloud water content and promote formation of precipitation and removal of condensate because ice crystals generally fall faster than liquid droplets (Mitchell et al., 2008; Eidhammer et al., 2017). Because this process can reduce cloud lifetime and cloud fraction, it can influence Earth’s radiative balance through aerosol-cloud interactions (Shi and Liu, 2019; Storelvmo, 2017).*”

Eidhammer, T., Morrison, H., Mitchell, D., Gettelman, A., and Erfani, E.: Improvements in

Global Climate Model Microphysics Using a Consistent Representation of Ice Particle Properties, *J. Climate*, 30, 609–629, <https://doi.org/10.1175/JCLI-D-16-0050.1>, 2017.

Mitchell, D. L., Rasch, P., Ivanova, D., McFarquhar, G., and Nousiainen, T.: Impact of small ice crystal assumptions on ice sedimentation rates in cirrus clouds and GCM simulations, *Geophys. Res. Lett.*, 35, L09806, <https://doi.org/10.1029/2008GL033552>, 2008.

Referee's comment 2-2:

Lines 41 – 43: Righi et al. (2025, ACP) may be relevant here since they show BC from aviation is not a significant INP.

Response:

Based on your comment, we have added Righi et al. (2025) to the sentence as follows (Lines 47–49): “Black carbon (BC), a product of incomplete combustion, has also been identified as a potential source of INPs, although its ice nucleating ability is lower than that of other aerosol species (Bond et al., 2013; Kanji et al., 2017; Righi et al., 2025).”

Righi, M., Testa, B., Beer, C. G., Hendricks, J., and Kanji, Z. A.: Aviation soot interactions with natural cirrus clouds are unlikely to have a significant impact on global climate, *Atmos. Chem. Phys.*, 25, 18341–18353, <https://doi.org/10.5194/acp-25-18341-2025>, 2025.

Referee's comment 2-3:

Table 1: Please add median particle size to Table 1. This will make it clearer that the greater mass concentration of dust translates to a higher INP number concentration. (For example, anomalously large INPs may dominate the mass concentration but not the number concentration.)

Response:

Based on your comment, we have added the particle sizes of aerosol types that act as INPs to Table 1 as follows:

Table 1. *Aerosol species and types acting as INPs in this study, and data sources for their emissions and ice nucleating abilities.*

<i>Aerosol species</i>	<i>Aerosol type</i>	<i>Emission</i>	<i>Ice nucleating ability</i>
<i>Dust</i>	<i>Arctic dust (39 nm–10 μm)</i>	<i>Online (Zender et al., 2003; Kok et al., 2014)</i>	<i>Tobo et al. (2019); Kawai et al. (2023)</i>
	<i>Non-Arctic dust (39 nm–10 μm)</i>		<i>DeMott et al. (2015)</i>
<i>Bioaerosols</i>	<i>Bacteria (1 μm)</i>		<i>Diehl and Mitra (2015)</i>
	<i>Fungal spores (5 μm)</i>	<i>Offline (Hoose et al., 2010)</i>	<i>Hummel et al. (2018)</i>
	<i>Pollen (30 μm)</i>		<i>Diehl and Mitra (2015)</i>
<i>MOA</i>	<i>MOA (0.2 μm)</i>	<i>Online (Burrows et al., 2013; Wilson et al., 2015)</i>	<i>Wilson et al. (2015)</i>
<i>BC</i>	<i>Biomass burning BC (39 nm–10 μm)</i>	<i>Dataset (van Marle et al., 2017)</i>	<i>Schill et al. (2020)</i>

In addition, the size distribution of emitted dust particles is based on Kok (2011). We explain this point in Section 2.1.1. as follows (Lines 122–123): “*The size distribution of emitted dust particles is based on Kok (2011).*” The size distribution of emitted BC particles is based on Matsui et al. (2022). We have added this point to Section 2.1.4 as follows (Lines 165–166): “*BC emissions are assumed to have number median diameters of 70 nm for anthropogenic sources and 100 nm for biofuel and biomass burning sources, with a geometric standard deviation of 1.8 (Matsui et al., 2022).*”

Matsui, H., Mori, T., Ohata, S., Moteki, N., Oshima, N., Goto-Azuma, K., Koike, M., and Kondo, Y.: Contrasting source contributions of Arctic black carbon to atmospheric concentrations, deposition flux, and atmospheric and snow radiative effects, *Atmos. Chem. Phys.*, 22, 8989–9009, <https://doi.org/10.5194/acp-22-8989-2022>, 2022.

Referee's comment 2-4:

Lines 89 – 91: Does this imply that the INP number concentration = the ice crystal number concentration (not the ice particle number concentration that is affected by aggregation)? More specifically, how are INPs defined? Do all INPs form ice crystals under favorable conditions? Or is there a probability, like 10%, that an INP will form an ice crystal under favorable conditions? If the former, then $\text{INP conc.} = \text{aerosol species \# conc.} \times \text{probability}$? In either case, please clearly define INP. It appears that Table 1 provides references for the INP probabilities used in this study. Please discuss how these probabilities are used.

Response:

In this study, INPs are defined as aerosol particles that can initiate ice formation under given thermodynamic conditions. The INP number concentration of each species is calculated by multiplying the mass or number concentration of each species by the activated fraction derived from its temperature-dependent ice nucleating ability. Therefore, not all aerosol particles act as INPs; rather, only a fraction of particles become active as INPs depending on temperature. We have added this explanation to Section 2.1 as follows (Lines 90–94): “*In other words, INPs are defined as aerosol particles that can initiate ice formation under given thermodynamic conditions. The INP number concentration of each species is calculated by multiplying the mass or number concentration of each species by the activated fraction derived from its temperature-dependent ice nucleating ability. Therefore, not all aerosol particles act as INPs, and only a fraction of particles become active as INPs depending on temperature.*”

The INP number concentration does not necessarily equal the ice crystal number concentration. In the cloud microphysics scheme, both water droplets and ice crystals are

prognostically treated in terms of their number concentrations and mixing ratios (Neale et al., 2012). Ice nucleation processes act to increase the ice crystal number concentration, and when the available INPs exceed the existing ice crystal number concentration, the model increases the ice crystal number concentration toward the INP number concentration. We have added this explanation to Section 2.1 as follows (Lines 98–104): *“In the cloud microphysics scheme, both water droplets and ice crystals are prognostically treated in terms of their number concentrations and mixing ratios (Neale et al., 2012). There are some ice formation pathways considered in the scheme (e.g., homogeneous freezing, immersion and condensation freezing, contact freezing, and secondary ice production), which change the number concentrations and mixing ratios of water droplets and ice crystals. The INP number concentration derived from the aerosol scheme contributes to the formation of ice crystals by increasing the number concentration of ice crystals. When the available INPs exceed the existing ice crystal number concentration, the model increases the ice crystal number concentration toward the INP number concentration.”*

Referee’s comment 2-5:

Lines 110 – 113: Roughly, what are the highest latitudes sampled by MODIS? This may inform the reader which latitudes have the highest confidence.

Response:

The highest latitudes sampled by MODIS are about 45° in winter and 80° in summer for each hemisphere. We have added this information to the sentence as follows (Lines 157–159): *“Missing values at high latitudes (about >45° in winter and >80° in summer) are imputed using the average north of 30° N for the Northern Hemisphere and south of 30° S for the Southern Hemisphere.”*

Referee’s comment 2-6:

Lines 119 – 121: Righi et al. (2025, ACP) also found that aviation soot has no significant impact on the INP concentration. Consider adding this reference above to help justify your practice of ignoring anthropogenic BC as an INP source.

Response:

As suggested, we have added Righi et al. (2025) to the sentence as follows (Lines 167–169): “*Because anthropogenic BC does not act efficiently as INPs (Kanji et al., 2017; Righi et al., 2025), this study does not consider its contribution as INPs.*”

Righi, M., Testa, B., Beer, C. G., Hendricks, J., and Kanji, Z. A.: Aviation soot interactions with natural cirrus clouds are unlikely to have a significant impact on global climate, Atmos. Chem. Phys., 25, 18341–18353, <https://doi.org/10.5194/acp-25-18341-2025>, 2025.

Referee’s comment 2-7:

Lines 132 – 135: Ice nucleating ability tends to decrease with increasing temperature, but here it abruptly increases moving from -9 °C to -5 °C. Was there a typo or is there a physical reason for this? If the latter, please provide the reason.

Response:

These values are scaling factors applied to the ice nucleating ability of bacteria used in the Base simulation. As shown in Fig. 1f (revised version), even within the suggested temperature range, the calculated ice nucleating ability of bacteria decreases with increasing temperature. We have clarified this point as follows (Lines 182–184): “*The ice nucleating ability of bacteria used in the Base simulation was multiplied by a scaling factor of $10^{3.2}$ at ≤ -35 °C, $10^{0.2}$ at -19 °C, $10^{-0.3}$ at -14 °C, $10^{-1.8}$ at -9 °C, and $10^{1.8}$ at -5 °C, with a lognormal interpolation between these temperatures.*”

Referee’s comment 2-8:

Lines 204 – 208: Same comment as for Lines 89 – 91.

Response:

Please see our Response to Referee’s comment 2-4.

Referee's comment 2-9:

Figure S8 in the Supplement: Panels marked Jan 2016 through Jul 2016 should be dated as 2017.

Response:

Thank you for pointing this out. We have corrected the year in Fig. S6 (revised version) as follows:

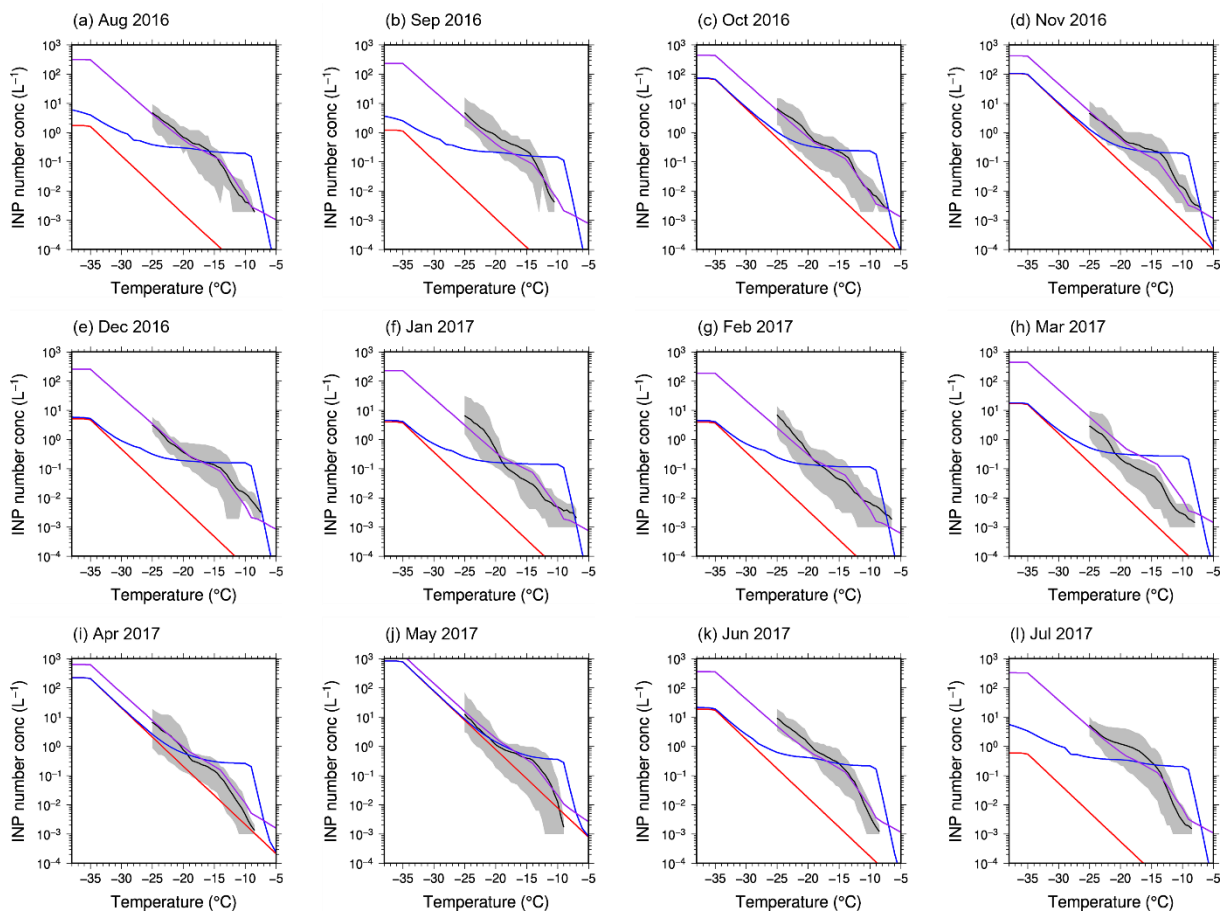


Figure S6. Monthly mean INP number concentrations as a function of freezing temperature observed at Tokyo Skytree, Japan, from August 2016 to July 2017 (Tobo et al., 2020) (black) and simulated for the corresponding location and period for dust only (red) and for all INP sources in the Base simulation (blue) and the observationally constrained simulation (purple). Gray shading indicates the range of observed INP number concentrations.

Response to Anonymous Referee #3

egusphere-2025-5850: “Global Modeling of Ice Nucleating Particles of Multiple Aerosol Species and Associated Cloud Radiative Effects” by K. Kawai, Z. Ren, and H. Matsui

Thank you very much for carefully reading our manuscript and providing valuable comments. We have revised the manuscript by taking your comments into account. Below, we describe our point-by-point responses to your comments. A revised manuscript with tracked changes has been uploaded.

Referee’s comment 3-1:

Aerosol bin microphysics: It is not clear to me, why a really complex aerosol scheme based on a bin approach is used in the investigation. Many other aerosol climate models use bulk model schemes to derive the global distributions of aerosols and INs. I think there are good reasons for using such a scheme, but this should be explained in the manuscript.

Response:

In this study, we used a bin scheme because it explicitly represents aerosol size distributions and microphysical processes, whereas bulk schemes rely on more simplified assumptions. This explicit treatment enables a more accurate simulation of aerosol mass and number concentrations. As described in the manuscript, the extensive evaluation of the model against various aerosol observations in our previous studies has demonstrated its reliability in reproducing aerosol concentrations, size distributions, and optical properties (Kawai et al., 2021a, 2021b, 2023; Kawai and Matsui, 2025; Liu et al., 2022, 2024a, 2024b; Matsui and Mahowald, 2017; Matsui et al., 2018a, 2018b, 2024). The use of the detailed aerosol scheme has substantially improved the model’s ability to reproduce long-range aerosol transport to remote regions, as well as aerosol mass and number concentrations in those regions (Liu and Matsui, 2021, 2022). This accuracy is particularly important in this study because the number concentrations of ice nucleating particles (INPs) strongly depend on aerosol mass and number concentrations. Therefore, the use of a bin scheme helps to reduce uncertainties associated with

aerosol representations and to improve the accuracy of INP estimates, which is essential for reliably assessing their impacts on clouds and radiation. We have added an explanation of this point to Section 2 as follows (Lines 72–82): “*We employed a sectional approach to explicitly represent aerosol size distributions and microphysical processes, enabling accurate simulations of aerosol mass and number concentrations. The extensive evaluation of the model against various aerosol observations in our previous studies has demonstrated its reliability in reproducing aerosol concentrations, size distributions, and optical properties (Kawai et al., 2021a, 2021b, 2023; Kawai and Matsui, 2025; Liu et al., 2022, 2024a, 2024b; Matsui and Mahowald, 2017; Matsui et al., 2018a, 2018b, 2024). The use of the detailed aerosol scheme has substantially improved the model’s ability to reproduce long-range aerosol transport to remote regions, as well as aerosol mass and number concentrations in those regions (Liu and Matsui, 2021, 2022). Such accuracy is essential for this study because INP number concentrations depend strongly on aerosol mass and number concentrations. The sectional approach therefore helps to reduce uncertainties in aerosol representations and to improve the reliability of estimates of INPs and their impacts on clouds and radiation.*”

Liu, M., and Matsui, H.: Improved simulations of global black carbon distributions by modifying wet scavenging processes in convective and mixed-phase clouds, J. Geophys. Res.-Atmos., 126, e2020JD033890, <https://doi.org/10.1029/2020JD033890>, 2021.

Liu, M., and Matsui, H.: Secondary organic aerosol formation regulates cloud condensation nuclei in the global remote troposphere, Geophys. Res. Lett., 49, e2022GL100543, <https://doi.org/10.1029/2022GL100543>, 2022.

Referee’s comment 3-2:

Coupling of aerosols and clouds schemes: Generally, the overall description of the aerosol and cloud models used in the GCM lacks details and is very short. While for the aerosol investigations a bin model is used, the cloud model (as far as I know the CAM model) is based on a bulk scheme. How do the properties of aerosols (in a bin scheme) translate into the cloud bulk scheme? The authors should explain the overall concept and give more details about the schemes. For instance, it is not clear if there is a discrimination in the ice modes by ice formation

pathways or if just one class for all ice nucleation pathways is used.

Response:

In the cloud microphysics scheme of CAM5, both water droplets and ice crystals are prognostically treated in terms of their number concentrations and mixing ratios (Neale et al., 2012). The INP number concentration derived from our aerosol scheme contributes to the formation of ice crystals by increasing the number concentration of ice crystals. Once ice crystals form, water droplets evaporate and transfer water vapor to the growing ice crystals via the Wegener-Bergeron-Findeisen process (Korolev, 2007). Through these interactions, aerosol properties represented by the bin scheme influence cloud microphysics and subsequent cloud evolution. There are some ice formation pathways in CAM-ATRAS (e.g., homogeneous freezing, immersion and condensation freezing, contact freezing, and secondary ice production). As suggested, we have added this explanation to Section 2.1 as follows (Lines 98–106): “*In the cloud microphysics scheme, both water droplets and ice crystals are prognostically treated in terms of their number concentrations and mixing ratios (Neale et al., 2012). There are some ice formation pathways considered in the scheme (e.g., homogeneous freezing, immersion and condensation freezing, contact freezing, and secondary ice production), which change the number concentrations and mixing ratios of water droplets and ice crystals. The INP number concentration derived from the aerosol scheme contributes to the formation of ice crystals by increasing the number concentration of ice crystals. When the available INPs exceed the existing ice crystal number concentration, the model increases the ice crystal number concentration toward the INP number concentration. Once ice crystals form, water droplets evaporate and transfer water vapor to the growing ice crystals via the Wegener-Bergeron-Findeisen process (Korolev, 2007). Through these interactions, aerosol properties represented by the sectional scheme influence cloud microphysics and subsequent cloud evolution.*”

Referee’s comment 3-3:

IN Parameterizations: There is not much information about the ice nucleation scheme; somewhere in the text it is mentioned that for the IN parameterizations a purely temperature dependent approach is used. This is somewhat surprising, since from the bin aerosol scheme

there should be much more information available in order to use a more detailed IN scheme. There is probably a reason for that, which should be stated in the manuscript.

Response:

INP parameterizations typically describe the temperature dependence of n_m and n_s (the ice nucleation active site density per unit mass or surface area, respectively). In this study, INP number concentrations are calculated using this temperature-dependent information together with the aerosol mass and number concentrations for each size bin simulated by the model. We have added this point to Section 2 as follows (Lines 95–97): “*INP parameterizations typically describe the temperature dependence of n_m or n_s (the ice nucleation active site density per unit mass or surface area, respectively). In this study, INP number concentrations are calculated using this temperature-dependent information together with the aerosol mass and number concentrations for each size bin simulated by the model.*”

Referee’s comment 3-4:

Fits for CRE (section 2.4): In the text the fit procedure is vaguely described with a reference to the supplement. Actually, just from the text this procedure cannot be understood in a meaningful way. I would recommend to include the figure (fig S2) into the main part of the manuscript.

Response:

As suggested, we have moved Fig. S2 to the main part of the manuscript (Fig. 2 in the revised version).

Referee’s comment 3-5:

Lack of MOA over China: From figure 1 one can conclude that there is no MOA contribution to INPs over China. Why is this so? Can this be explained by the wind fields (no transport of marine air to the continent) or is it just because the dust contribution is so large that the others cannot be seen? For figure 1 and 3 it would be good to have the numbers (including the fraction of contributions) in a separate table.

Response:

As the reviewer suggested, the contribution of MOA to the INP observations in China is very small, probably because MOA is not efficiently transported into the continental interior due to the wind fields (Fig. S2c in the revised version), and dust concentrations are high due to the proximity of a dust source region (i.e., the Gobi Desert) (Fig. S3 in the revised version). We have added this explanation to Section 3.1 as follows (Lines 225–227): “*For the observations in China, the contributions of MOA to total INPs are very small because MOA is not efficiently transported into the continental interior because of the prevailing wind fields (Fig. S1) and the dust concentrations are high because of the proximity of a dust source region (i.e., the Gobi Desert) (Fig. S2).*”

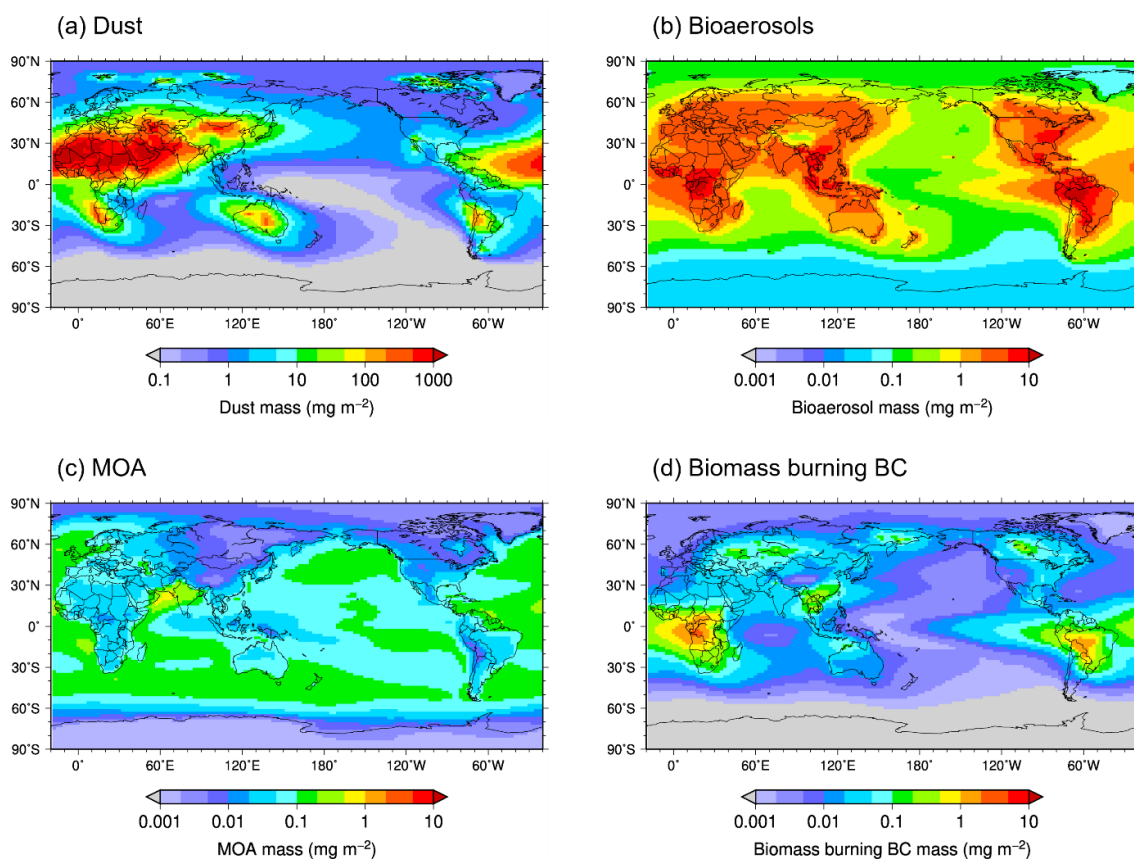


Figure S1. Annual mean vertically integrated mass concentrations of (a) dust, (b) bioaerosols, (c) MOA, and (d) biomass burning BC.

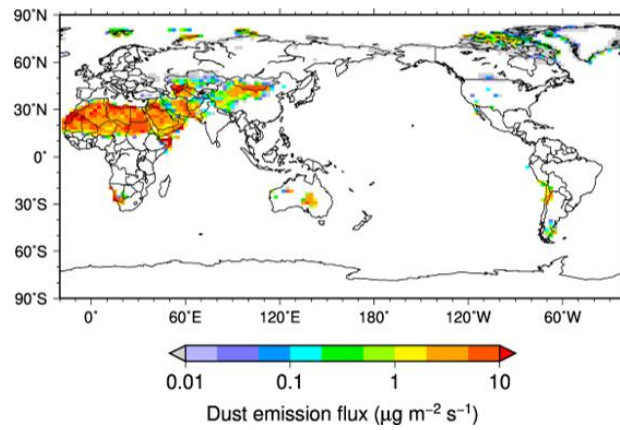


Figure S2. Annual mean dust emission fluxes simulated by the model.

In addition, as suggested, we have added Tables S1 and S2, which show the percentages of the aerosol species in total INPs in Figs. 3 and 5 (revised version), respectively, to Supplements as follows:

Table S1. Fractions of dust, bioaerosols, MOA, and BC in the simulated total INP number concentrations for the observations at Beijing, China (Hu et al., 2023); Vancouver Island, Canada; Saclay, France (Mason et al., 2016); and Svalbard, Arctic (March 2017) (Tobo et al., 2019) at different freezing temperatures.

	Temperature (°C)	Dust (%)	Bioaerosols (%)	MOA (%)	BC (%)
China	−20	87	13	0.011	0.0065
	−15	41	59	0.0056	0.0036
	−8	23	77	0.0035	0.0
Canada	−25	0.22	68	31	0.31
	−20	0.032	95	4.8	0.052
	−15	0.011	98	1.6	0.019
France	−25	24	73	3.2	0.12
Arctic	−25	0.45	1.9	98	0.0044
	−20	0.37	15	85	0.00076

–16 0.20 51 49 0.00026

Table S2. *Fractions of dust, bioaerosols, MOA, and BC in the annual and global mean vertically integrated total INP number concentrations in the whole atmosphere (2–1000 hPa) and in the upper (100–400 hPa), middle (400–700 hPa), and lower troposphere (700–1000 hPa).*

	Dust (%)	Bioaerosols (%)	MOA (%)	BC (%)
Whole atmosphere	97	2.4	0.52	0.11
Upper troposphere	98	1.7	0.33	0.16
Middle troposphere	96	3.1	0.79	0.032
Lower troposphere	60	37	3.2	0.029

Referee’s comment 3-6:

Aerosol distribution in the upper levels: It seems that for the upper levels (i.e. at cold temperatures of $T \sim -25^{\circ}\text{C}$, see comparison with Tokyo) the agreement with the observations is less convincing. Can you give some (physical/chemical/meteorological) reasons for that?

Response:

For the comparisons with the observations at Tokyo (Fig. 4 in the revised version), the simulated INP number concentrations were calculated from the given temperature and the simulated aerosol number or mass concentrations at the tower observation site (not the upper levels) using the ice nucleating abilities of the various aerosol species. At -25°C , the model still underestimates the observations by about one order of magnitude during summer and winter. This discrepancy might be attributable to uncertainties in dust emission and transport or to unknown local sources of INPs (e.g., local soil dust). We explain this point in Section 3.1 as follows (Lines 233–235): “At -25°C , the model still underestimates the observations by about

one order of magnitude during summer and winter. This discrepancy might be attributable to uncertainties in dust emission and transport or to unknown local sources of INPs (e.g., local soil dust)."

In addition, Liu and Matsui (2021, 2022) demonstrated that the model has a high ability to reproduce observed aerosol mass and number concentrations in the upper troposphere over remote regions. We have added this point to Section 2 (Lines 78–79) as follows: "*The use of the detailed aerosol scheme has substantially improved the model's ability to reproduce long-range aerosol transport to remote regions, as well as aerosol mass and number concentrations in those regions (Liu and Matsui, 2021, 2022).*"