

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).*”

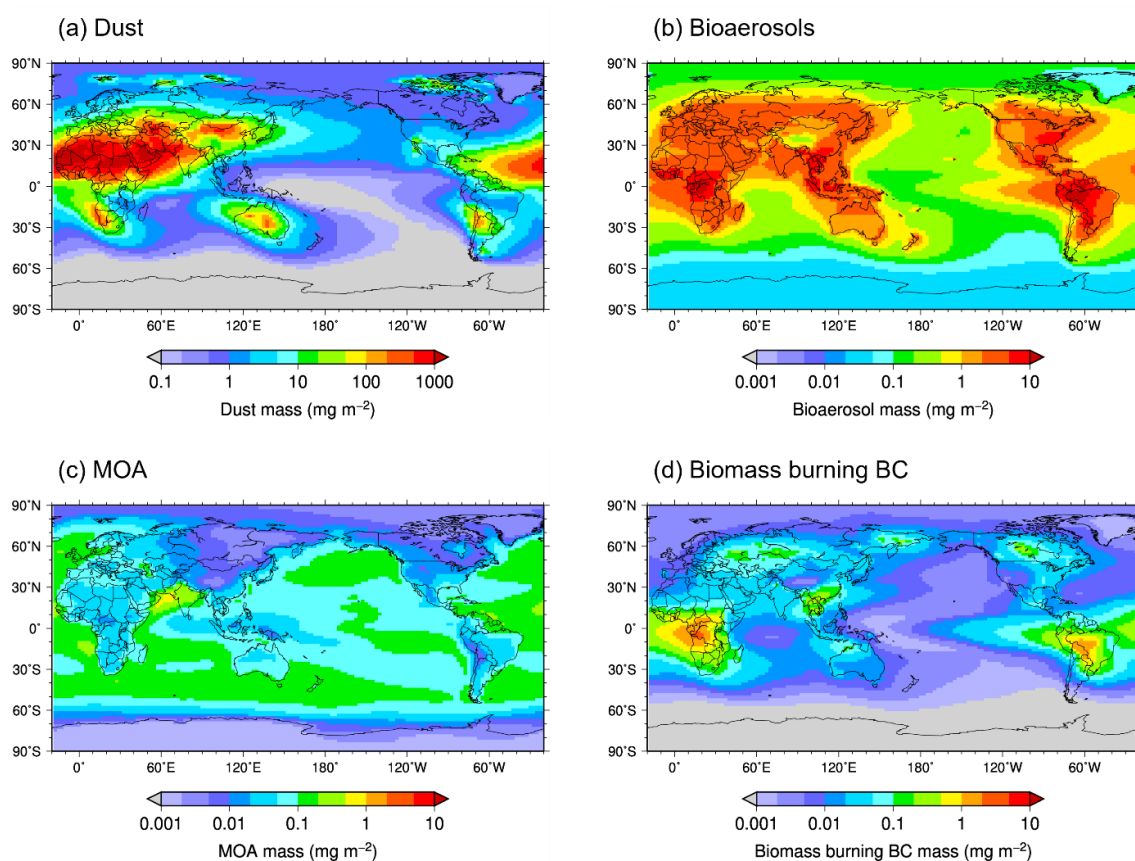


Figure S1. Annual mean vertically integrated mass concentrations of (a) dust, (b) bioaerosols, (c) MOA, and (d) biomass burning BC.

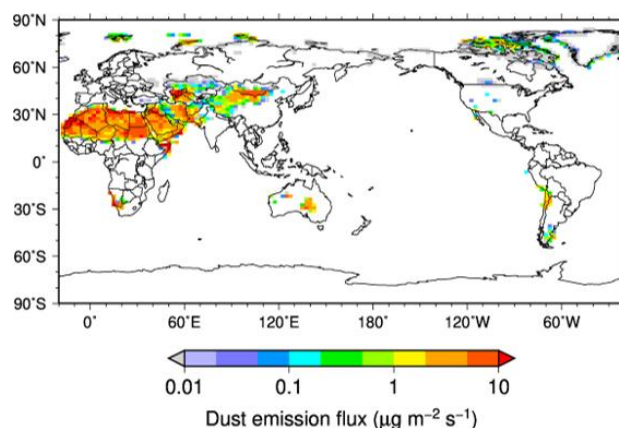


Figure S2. Annual mean dust emission fluxes simulated by the model.

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 (%)
<i>China</i>	−20	87	13	0.011	0.0065
	−15	41	59	0.0056	0.0036
	−8	23	77	0.0035	0.0
<i>Canada</i>	−25	0.22	68	31	0.31
	−20	0.032	95	4.8	0.052
	−15	0.011	98	1.6	0.019
<i>France</i>	−25	24	73	3.2	0.12
<i>Arctic</i>	−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~ -25°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).*”