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Nov 19th, 2025

Dear ACP Reviewer,

Thank you for your feedback and careful reading of our manuscript. We are happy to submit a revised manuscript, "*Resolving the roles of soot and dust in cirrus cloud ice formation at regional and global scales: insights from parcel and climate models*". We highly appreciate your time and valuable suggestions. Below, we provide detailed responses to all reviewer comments. For clarity, the reviewer comments are presented in **gray**, our responses in **black**, and revised text in **blue**. We hope that our revisions address all concerns.

Best wishes,
Xiaohan Li, Songmiao Fan, Huan Guo, Paul Ginoux

Summary of Content:

Reply to Reviewer 2

Reviewer: 2

This study uses a parcel model to find relationships on the competition between homogeneous and heterogeneous nucleation based upon a large variety of black carbon and dust concentration. The resulting findings are inserted into the GFDL AM4-MG2 model to examine how black carbon and mineral dust impacts cirrus clouds globally. Black carbon enhances global ice crystal number concentration and significantly increases the annual global longwave cloud radiative effect, causing warming, particularly during polar winters. The results emphasize the roles of soot and dust and the need to assess the climate impact of increasing wildfire emissions on atmospheric ice processes. The paper is very well written, and conclusions are well supported. I have only minor comments.

Reply to reviewer #2 summary. Thanks for your careful reading of our manuscript. We deeply appreciate your valuable comments and suggestions. Below you will find our replies to your comments.

In the abstract, line 8: I like to reserve the word observe to real observations. I wonder if the sentence could be rephrased to something like "The strongest enhancements are found during boreal"

Page 5, line 147: Add "of" ….where N_aer is the number density of ice nucleating….

Page 7, line 209. ICNC has only been defined in the abstract, but not in the main text yet.

Reply to Q1: Thanks for raising the above comments. We have revised the text as suggested and added the definition of ICNC (ice crystal number concentration) in the last paragraph of the Introduction to ensure clarity for readers.

Page 4, lines 118-123: The diameters and standard deviations of the aerosol size distribution are fixed. Could varying the size and shape of the size distributions change your results?

Reply to Q2: We thank the reviewer for this insightful question. Indeed, varying the size and shape of the aerosol size distribution could potentially influence the results. However, as described in the methodology, AM4-MG2 employs a bulk aerosol scheme in which only the aerosol mass concentration is prognosed with the mean particle size and geometric standard deviation are fixed and prescribed. This assumption of fixed aerosol size and distribution is a common practice in GCM simulations, including those participating in CMIP5 and CMIP6. We acknowledge that aerosol size can influence activation and ice nucleation processes, but given the large uncertainties in observed size distributions and to maintain consistency with GCM representations, our parcel model experiments also adopt prescribed, fixed size parameters. The effects of varying aerosol size can be investigated once a fully coupled aerosol microphysics scheme becomes available, which is currently under active development at GFDL. We have added the following text in the second paragraph of Section 2.1.1 to acknowledge the impact of size distribution and limitations:

"We note that a fixed aerosol size distribution is used in this study, although varying the size and shape of the distribution could potentially influence the results. The choice of fixed size parameters is primarily to maintain consistency with the bulk aerosol scheme in the host climate model AM4-MG2, where only the aerosol mass concentration is prognosed, and the mean particle size and geometric standard deviation are prescribed. This bulk representation is a common practice in GCM simulations, including those participating in CMIP5 and CMIP6. We acknowledge that aerosol size can influence activation and ice nucleation processes; however, given the large uncertainties in observed size distributions and to ensure consistency with GCM representations, our parcel model experiments also adopt prescribed, fixed size parameters. The effects of varying aerosol size will be explored in future studies once a fully coupled aerosol microphysics scheme becomes available, which is currently under active development."

Page 7, line 199. It is stated that the AMIP simulation is run through 2006. Why is the analysis period only to 2005 and not through 2006?

Reply to Q3: Thank you for pointing this out. The AMIP simulation was initialized in 2000 and extended into early 2006 only to generate model restart files. Therefore, the complete simulation years available for analysis are from 2000 to 2005. To avoid confusion, we have revised the text as follows:

"The simulation was initialized in 2000 and run to the end of 2006, with the first year treated as model spin-up, and the following 5 years of 2001–2005 for analysis."

Section 3.1.2 You describe the ICNC dependence on aerosol composition. But there is no mention about the competition between homogeneous and heterogeneous dependence on aerosol composition. I am not sure how to present it, but it would be interesting to have a figure showing how the change in composition impacts homogeneous freezing.

Reply to Q4: Homogeneous ice nucleation is not entirely negligible in our parcel model. We do observe homogeneous nucleation under warmer cirrus conditions (typically for temperatures above ~230 K) and when dust and BC concentrations are sufficiently low.

To illustrate the role of homogeneous nucleation, we computed the fraction of homogeneous ice crystals, defined as

$$f_{\mathrm{homo}} = \frac{N_{i,\mathrm{tot}} - N_{i,\mathrm{dust}} - N_{i,\mathrm{BC}}}{N_{i,\mathrm{tot}}}$$

and plotted it as a function of updraft velocity at 232 K and 300 hPa, using sea salt and sulfate concentrations of 100 ng m-3 for various combinations of dust and BC mass concentrations (new Figure A10). Figure A10 shows that when dust and BC concentrations are low, homogeneous nucleation contributes up to ~96% of total ice crystals. As expected, this fraction decreases as INP concentrations increase (from left to right panels).

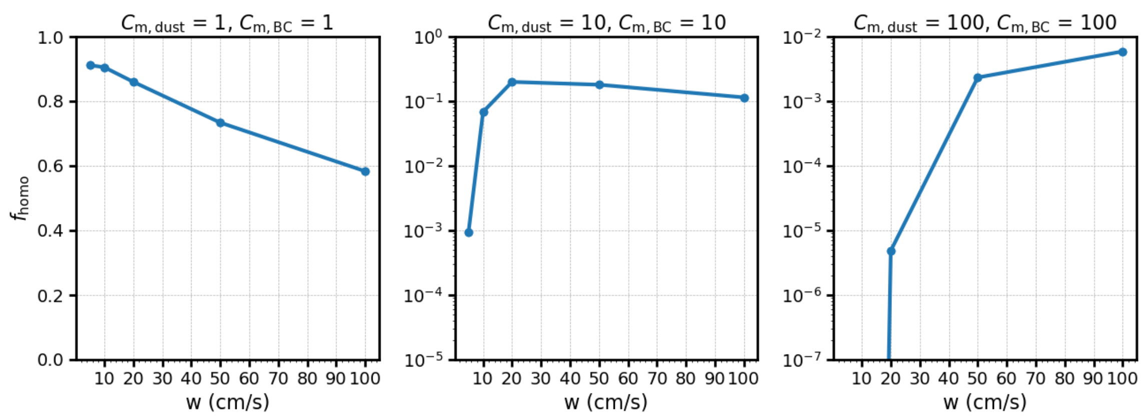

Figure A3: Fraction of ice crystals formed by homogeneous nucleation f_homo as a function of updraft velocity w at 232 K and 300 hPa, with sulfate and sea-salt concentrations fixed at 100 ng/m-3. Panels show different combinations of dust and BC mass concentrations in unit of ng/m^{-3} (from low to high, left to right). When dust and BC concentrations are low, homogeneous nucleation contributes up to more than 90% of total ice crystals. The contribution decreases with increasing INP concentrations at the same updraft velocity, consistent with the expected suppression of homogeneous nucleation by heterogeneous nucleation on dust and soot.

To illustrate the temperature dependence, we also plotted the maximum value of f_{homo} across all updraft velocities as a function of temperature (new Figure A11). Figure A11 demonstrates that homogeneous nucleation becomes important only at warmer temperatures (typically $T \gtrsim 225$ K). At colder temperatures, the maximum homogeneous fraction is very small (<1%), consistent with the dominance of heterogeneous nucleation on dust and soot at these conditions.
These additions clarify that homogeneous nucleation does occur in our parcel model, but its contribution becomes negligible at colder cirrus temperatures due to the rapid consumption of supersaturation by heterogeneous ice formation and growth.

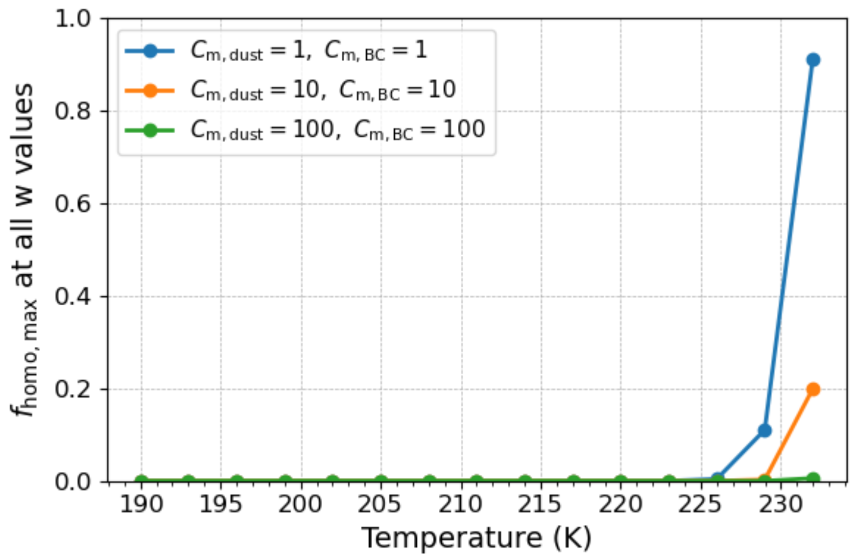

Figure A4: Maximum fraction of homogeneous ice crystals f_{homo} across all simulated updraft velocities as a function of temperature. Homogeneous nucleation becomes important only at warmer temperatures (typically $T \gtrsim 225$ K), where the maximum value of f_{homo} can be substantial. At colder temperatures, the homogeneous fraction is negligible (<1%), reflecting the strong depletion of supersaturation by heterogeneous nucleation on dust and soot. The different color lines show different combinations of dust and BC mass concentrations in unit of ng/m^{-3}

To clarify this competition between homogeneous and heterogeneous nucleation, we have added the above two Figures to the manuscript, and added the discussion in the first paragraph of Section 3.1.2 as:
"We note that homogeneous nucleation is not completely negligible in our parcel model. Our results show that under warmer cirrus conditions (typically for temperatures above 230 K) and when dust and BC concentrations are low, the fraction of ice crystals formed by homogeneous nucleation, defined as f_homo = (N_i,tot-N_i,dust-N_i,BC)/N_i,tot, can reach values as high as ~96%. The dependence of f_homo on INP concentration, temperature, and updraft velocity is shown in Figures A3 and A4."

Page 11, Figure 3. The In-situ Heymsfield symbols are difficult to see, special in the green region. The dotted lines could also be slightly enhanced. It would also be interesting to see how the model performs without the BC in this figure.
Reply to Q5: Thank you for this comment. We have reduced the transparency of the in-situ Heymsfield observations so that the symbols are more visible, particularly in regions with dense model output. We have also slightly enhanced the dotted lines for improved clarity.

Regarding the suggestion to include the model results without BC directly in Figure 3, we appreciate the idea; however, the background density map differs substantially between the BC and no-BC simulations because the underlying ICNC distributions shift in magnitude and structure. Overlaying the two density fields in the same panel would therefore not allow a meaningful or visually interpretable comparison. Instead, as shown in the Appendix, we provide a figure displaying the difference in ICNC between the BC and no-BC simulations as a function of temperature. This allows a clear comparison of the BC contribution while avoiding overlapping density biases.

Conclusion: Lines 480-485. The authors have based their conclusion on one dust and BC parameterization. There are other parameterizations out there that can lead to different results. For example, how is the temperature dependent active site density in other parameterizations and how could this impact the results?
Reply to Q6: We thank the reviewer for highlighting this important point regarding the uncertainties associated with different ice nucleation parameterizations. We agree that using alternative formulations could influence the results and

we plan to collaborate with others to compare different parameterizations and assess their impacts on simulated cirrus properties. We also note that the parameterization from Ulrich et al, 2017, used in this study, is based on multiple dust and soot samples measured in the AIDA chamber, thus providing a representative, though not exhaustive dataset. We have added the following sentences in the last paragraph of the conclusion part to acknowledge the uncertainties arising from different parameterizations:
"It should also be noted that other parameterizations for the ice-nucleating ability of dust and soot exist beyond those applied in this study, and alternative formulations may yield different results. Future collaborative efforts to intercompare parameterizations and quantify their impacts on simulated cirrus properties would therefore be valuable. "

Figures 5 and 8 are very small and hard to read on printed paper.
Reply to Q7: Thank you for pointing this out. We have updated Figures 5 and 8 by increasing their size and adjusting the layout to improve readability, especially when printed.