



Supplement of

**Prognostic simulations of mixed-phase clouds with model AC-1D v1.0:
the impact of aerosol types and freezing parameterizations on ice crystal
budgets**

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Table S1: Activated fraction of the total aerosol reservoir over time at the average cloud temperature 254.15 K (-19.0 °C).

Aerosol Type	τ_{act}	Activated Fraction (1 h)	Activated Fraction (10 h)
Mineral Dust	1.08×10^5 s	3.3%	28.3%
Organic	$> 10^7$ s	$< 0.01\%$	$< 0.1\%$
SSA	$> 10^6$ s	$< 0.1\%$	$< 1.0\%$

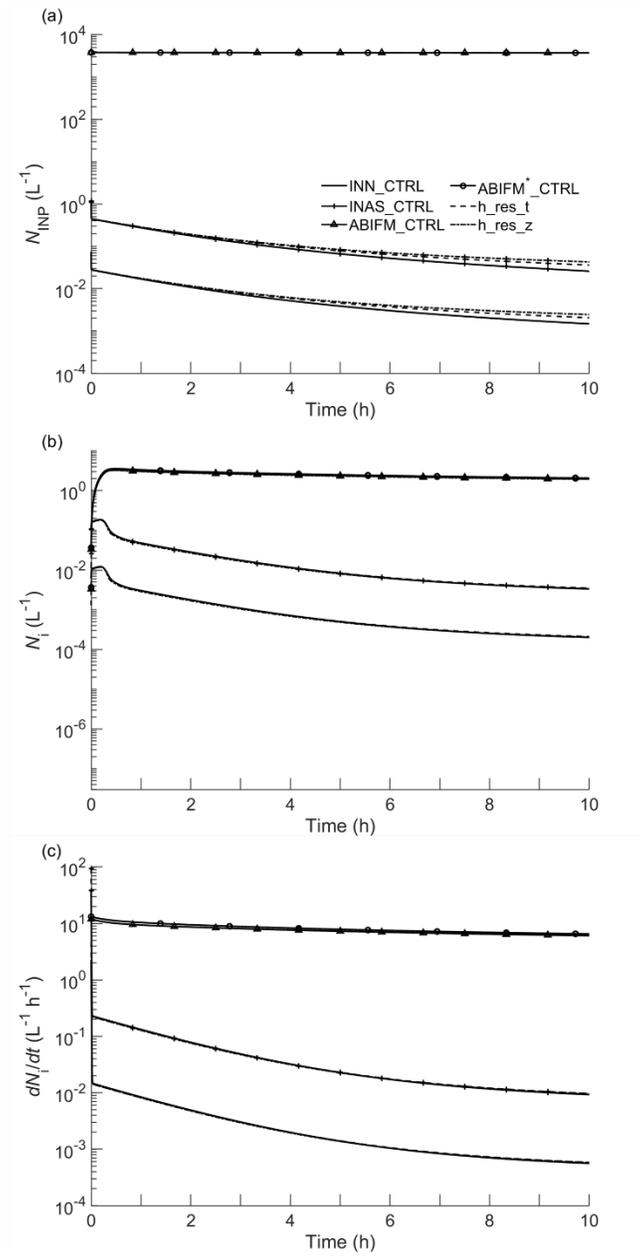


Figure S1. Time series of simulated domain averaged (a) activatable INP number concentration (N_{INP}), (b) ice crystal number concentration (N_i), and (c) ice crystal formation rate (dN_i/dt) while applying a time step length (δt) of 1 s (dash-dotted line) and a vertical resolution (δz) of 5 m (dashed line). Simulations are all initialized with the same PSD for dust and cloud parameters with the original values. Simulation results include different immersion freezing parameterizations: INN (no symbols), INAS (cross), ABIFM (triangles), ABIFM* (circles). In all panels, the solid lines indicate simulation results using the original time step and vertical resolution.

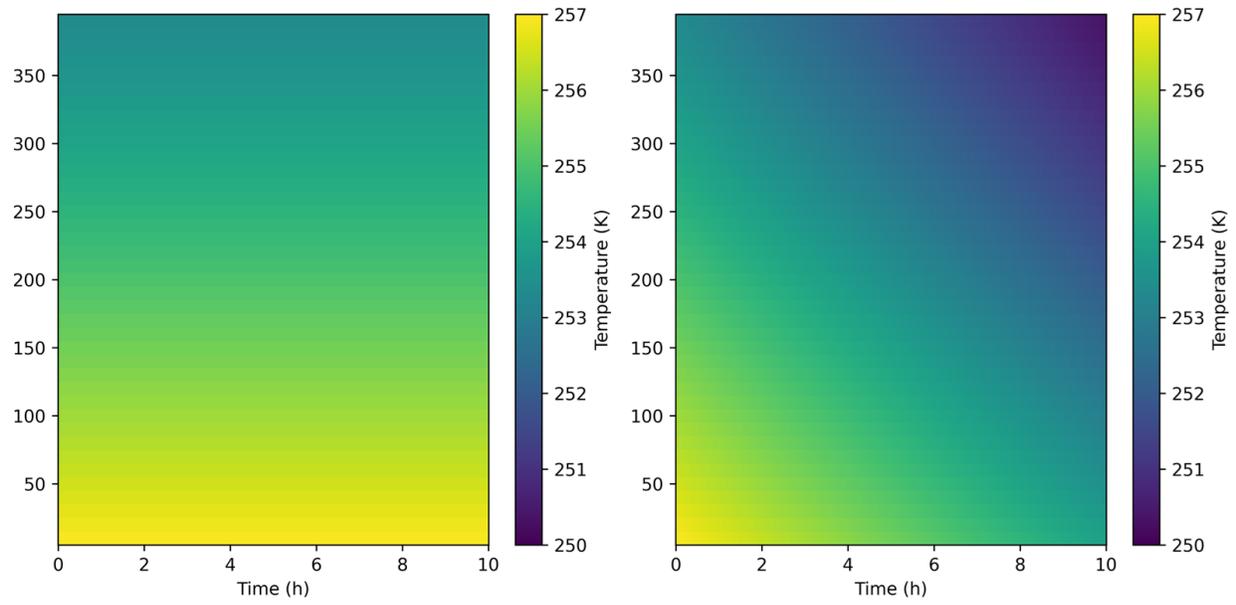


Figure S2. The evolution of temperature for respective case studies. (A) Control run (CTRL) (initial temperature profile without cloud cooling rate (CCR = 0)). (B) CCR=0.3 (initial temperature profile with CCR=0.3 °C h⁻¹ over the whole domain).

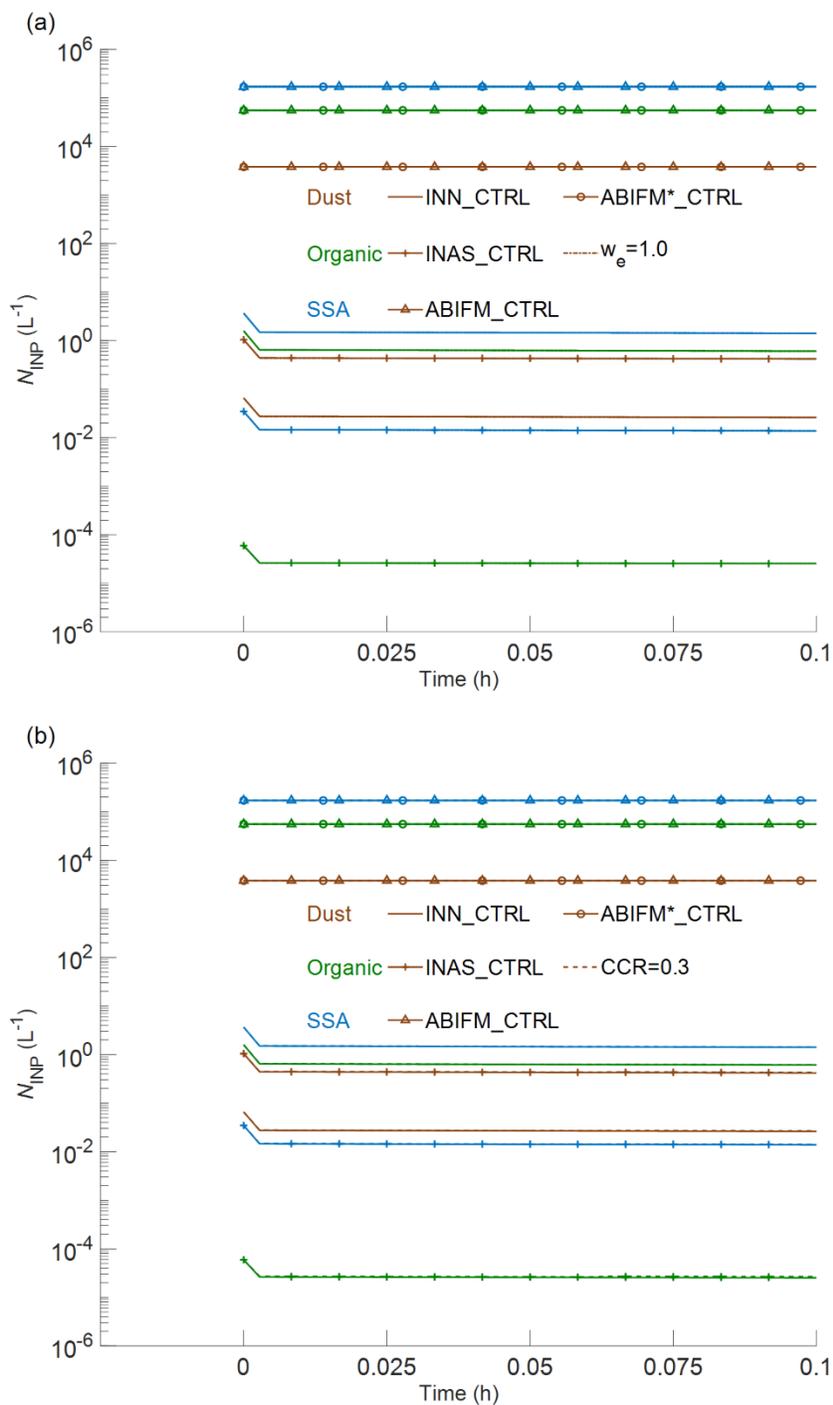


Figure S3. Time series of simulated domain-averaged activatable INP number concentration (N_{INP} in L^{-1}) for beginning 0.1 hours when changing the cloud-top entrainment rate (a) and cloud cooling rate (b). Simulations are initialized with different aerosol PSDs (dust, organic, and SSA particles), immersion freezing parameterizations (INN, INAS, ABIFM, ABIFM*) and cloud parameters (cloud cooling rate, cloud-top entrainment rate). Brown, green, and blue lines represent the application of aerosol PSDs of dust, organic, and SSA particles, respectively. Simulation results represent different immersion freezing parameterizations: INN (no symbols), INAS (cross), ABIFM (triangle), and ABIFM* (circle). In both panels, the thin solid lines indicate results with the original, unperturbed cloud parameters (CTRL). The dashed lines denote results with the cloud cooling rate (CCR) of $0.3 \text{ } ^\circ\text{C h}^{-1}$ (CCR = 0.3) and the dash-dotted lines show the results with the cloud-top entrainment rate (w_e) of 1 cm s^{-1} ($w_e = 1.0$).

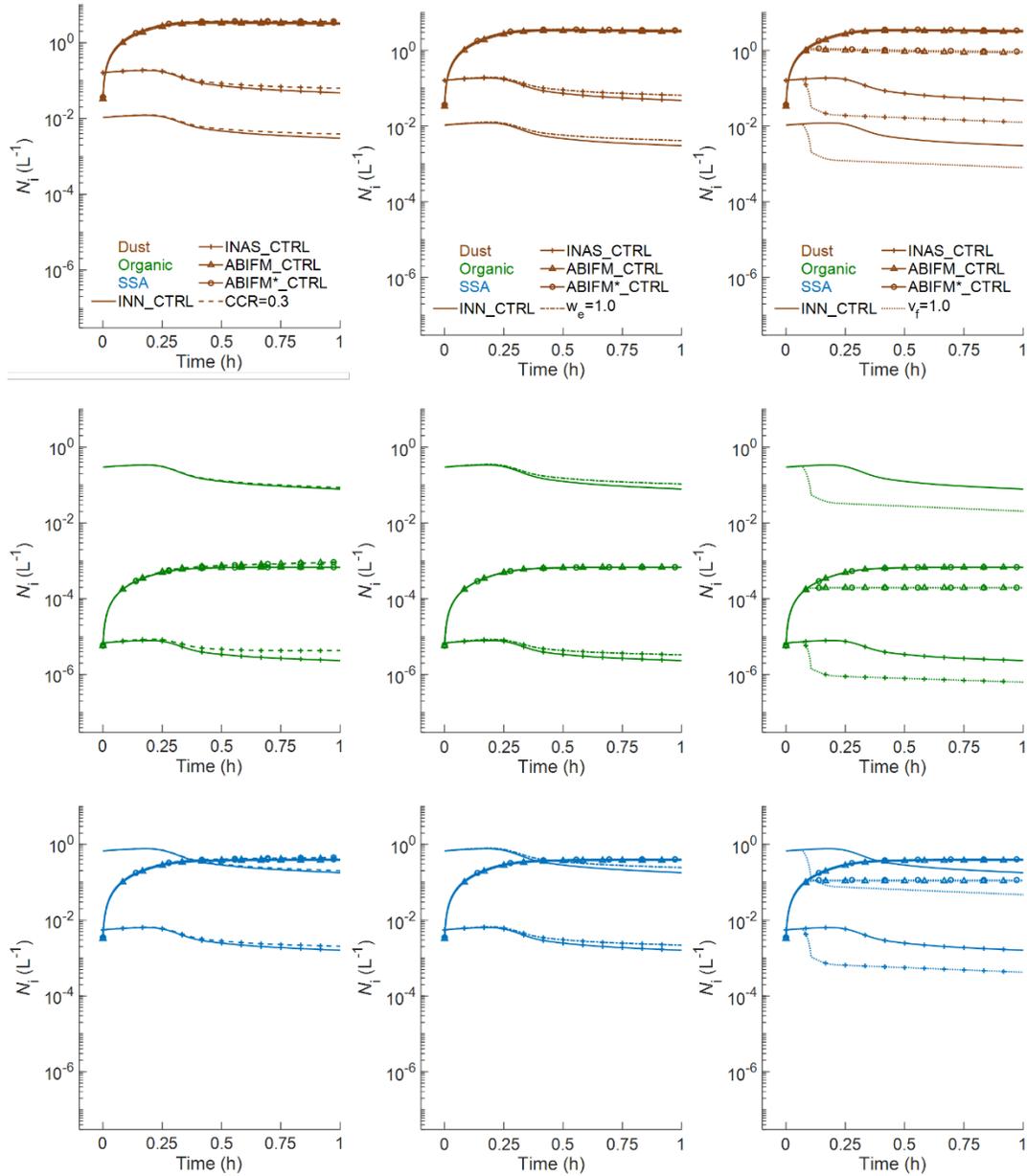


Figure S4: Temporal evolution of the domain-averaged ice crystal number concentration (N_i in L^{-1}) for the initial 1 hour in response to different cloud system parameters. The nine panels are organized by aerosol type in rows (mineral dust, top; organic, middle; and sea spray aerosol (SSA), bottom) and by sensitivity experiment in columns. The columns from left to right represent simulations with an applied cloud cooling rate (CCR), an enhanced entrainment rate (w_e), and an increased ice crystal fall speed (v_f), respectively. Within each panel, different line styles and colors represent the four immersion freezing (IMF) parameterizations, with legends and styling identical to those used in Figure S3.

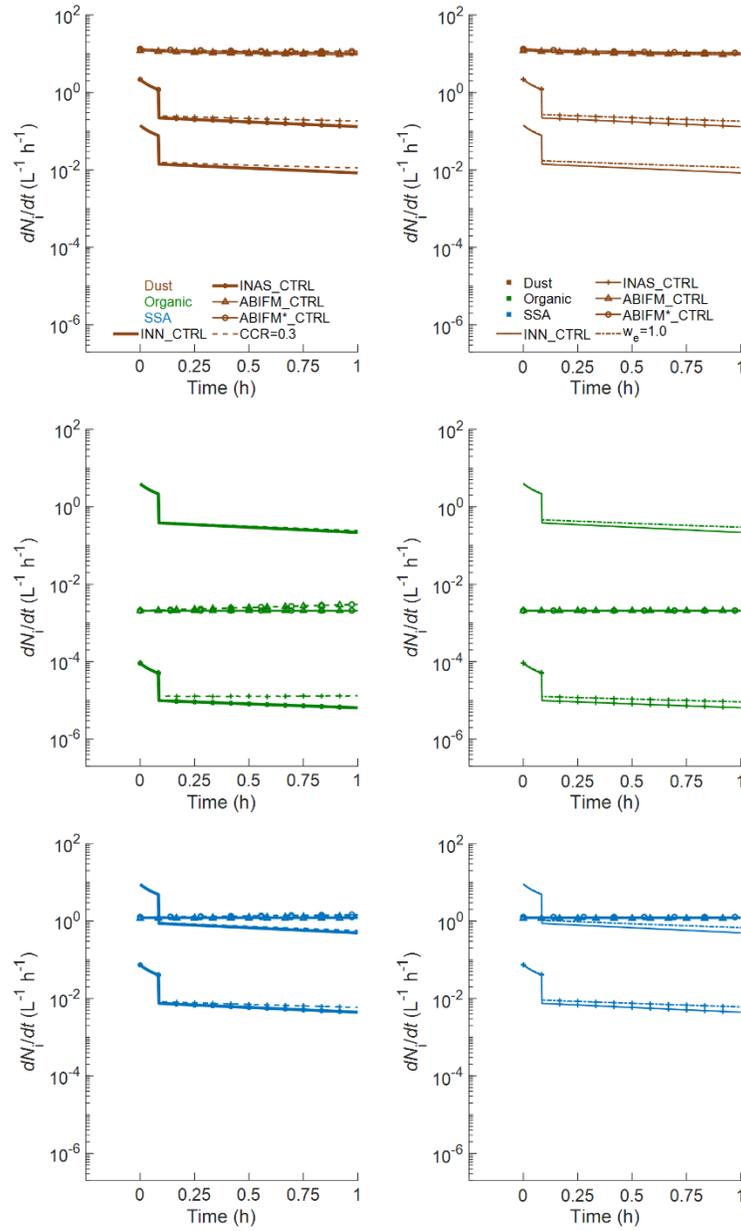


Figure S5: Temporal evolution of the domain-averaged ice crystal formation rate (dN_i/dt in $L^{-1} h^{-1}$) for the initial 1 hour. The six panels are organized by aerosol types in rows (mineral dust, top; organic, middle; and sea spray aerosol (SSA), bottom) and by sensitivity experiment in columns. The columns from left to right show the response to an applied cloud cooling rate (CCR), and enhanced entrainment rate (w_e). Legends and line styles for the four immersion freezing (IMF) parameterizations are identical to those used in Figure S4.

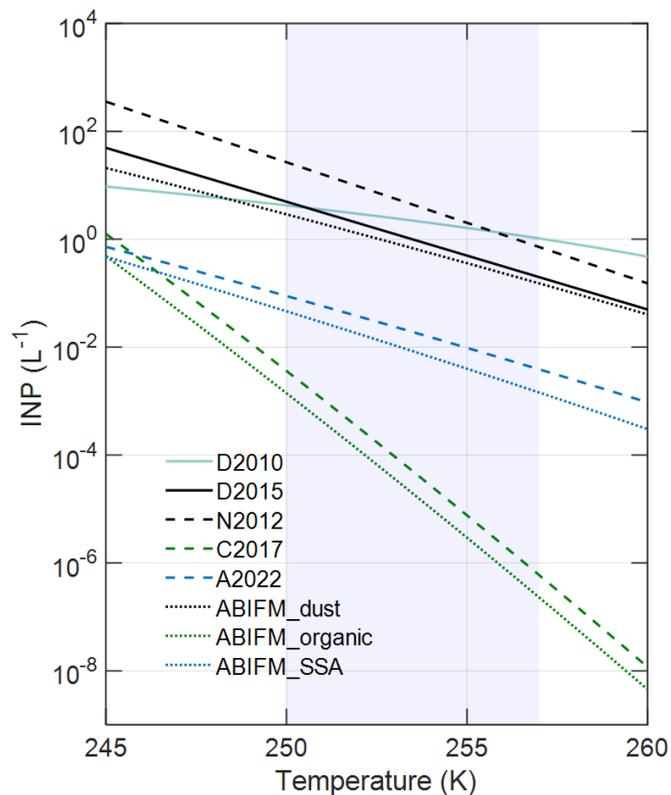


Figure S6. The predicted number concentration of INP using the same PSD for respective immersion freezing parameterizations, INN (solid lines), INAS (dashed lines), ABIFM (dotted lines). The chosen activation time for ABIFM is 1 min following the recommendation given in Alpert et al. (2022). Black, blue, and green lines represent the application of dust, organic, and SSA particles. Light blue represents ambient particles. Detailed information is summarized in Table 3. The blue shading indicates the temperature range in the simulation domain. The immersion freezing parameterizations are originally obtained from DeMott et al. (2010), DeMott et al. (2015), Niemand et al. (2012), China et al. (2017), Alpert et al. (2022), and (Knopf and Alpert, 2013).

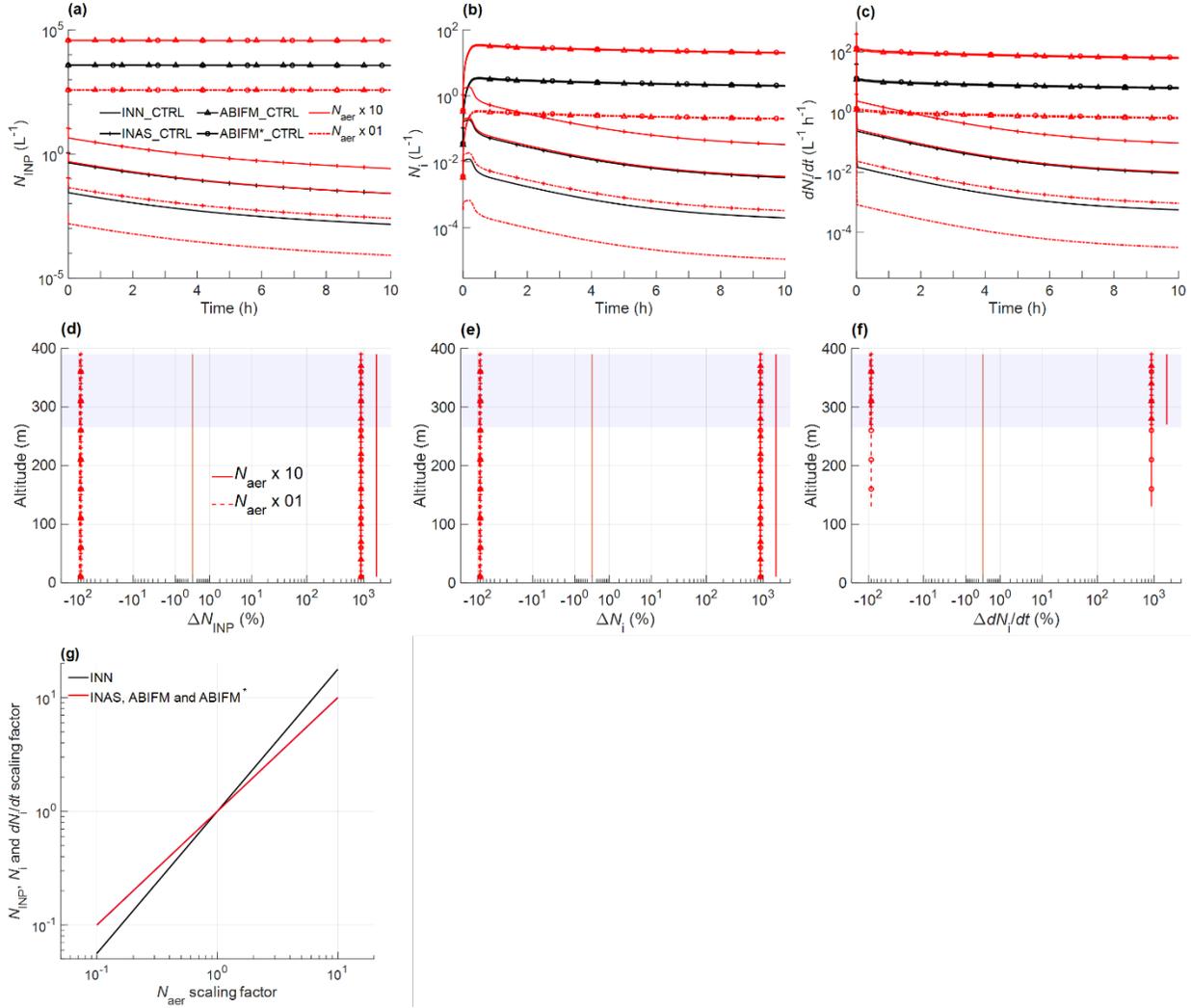


Figure S7. Results of the sensitivity tests involved changing the aerosol number concentration ($N_{\text{aer}} \times 10$, increasing aerosol concentration by a factor of 10 and $N_{\text{aer}} \times 01$, decreasing aerosol concentration by a factor of 10). (a, b, c) Time series of simulated domain averaged activatable INP number concentration (N_{INP}), ice crystal number concentration (N_i), and ice crystal formation rate (dN_i/dt) while changing the N_{aer} , respectively. (d, e, f) Vertical profiles of the change in number concentration of INP (ΔN_{INP}), number concentration of ice crystal (ΔN_i), and ice crystal formation rate ($\Delta dN_i/dt$), respectively (averaged over entire 10 h of simulation time). Simulation results include immersion freezing parameterizations: INN (no symbols), INAS (cross), ABIFM (triangles), ABIFM* (circles). Black solid lines, red solid lines and red dashed lines represent CTRL, $N_{\text{aer}} \times 10$, and $N_{\text{aer}} \times 01$, respectively. The blue shaded area denotes the cloud layer and the vertical orange lines in the center of x-axis highlight the value of 0. (g) Relation between cloud properties (N_{INP} , N_i , and dN_i/dt) scaling factor and N_{aer} scaling factor. Black solid and red solid lines represent simulation with INN and with INAS, ABIFM, and ABIFM*, respectively. Notice that simulations are initialized with the same PSD for dust particles only.

References

- Alpert, P. A., Kilhau, W. P., O'Brien, R. E., Moffet, R. C., Gilles, M. K., Wang, B., Laskin, A., Aller, J. Y., and Knopf, D. A.: Ice-nucleating agents in sea spray aerosol identified and quantified with a holistic multimodal freezing model, *Sci. Adv.*, 8, eabq6842, <https://doi.org/10.1126/sciadv.abq6842>, 2022.
- China, S., Alpert, P. A., Zhang, B., Schum, S., Dzepina, K., Wright, K., Owen, R. C., Fialho, P., Mazzoleni, L. R., Mazzoleni, C., and Knopf, D. A.: Ice cloud formation potential by free tropospheric particles from long-range transport over the Northern Atlantic Ocean, *J. Geophys. Res.-Atmos.*, 122, 3065-3079, <https://doi.org/10.1002/2016jd025817>, 2017.
- DeMott, P. J., Prenni, A. J., Liu, X., Kreidenweis, S. M., Petters, M. D., Twohy, C. H., Richardson, M. S., Eidhammer, T., and Rogers, D. C.: Predicting global atmospheric ice nuclei distributions and their impacts on climate, *Proc. Nat. Acad. Sci.*, 107, 11217-11222, <https://doi.org/10.1073/pnas.0910818107>, 2010.
- DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., Niemand, M., Möhler, O., Snider, J. R., Wang, Z., and Kreidenweis, S. M.: Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles, *Atmos. Chem. Phys.*, 15, 393-409, <https://doi.org/10.5194/acp-15-393-2015>, 2015.
- Knopf, D. A. and Alpert, P. A.: A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets, *Faraday Discuss.*, 165, 513-534, <https://doi.org/10.1039/c3fd00035d>, 2013.
- Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., and Leisner, T.: A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles, *J. Atmos. Sci.*, 69, 3077-3092, <https://doi.org/10.1175/Jas-D-11-0249.1>, 2012.