Supplement of Competition for water vapour results in suppression of ice formation in mixed phase clouds

Emma L. Simpson, Paul J. Connolly, and Gordon B. McFiggans

Centre for Atmospheric Science, School of Earth and Environmental Sciences, University of Manchester, Manchester, M13 9PL, UK

Correspondence to: Emma Simpson (emma.simpson@manchester.ac.uk

1 Sensitivities to model bin structure

The fraction of frozen INP plotted against the diameter of INP, Supplementary Fig. S1., reveals sensitivities to the bin structure used.

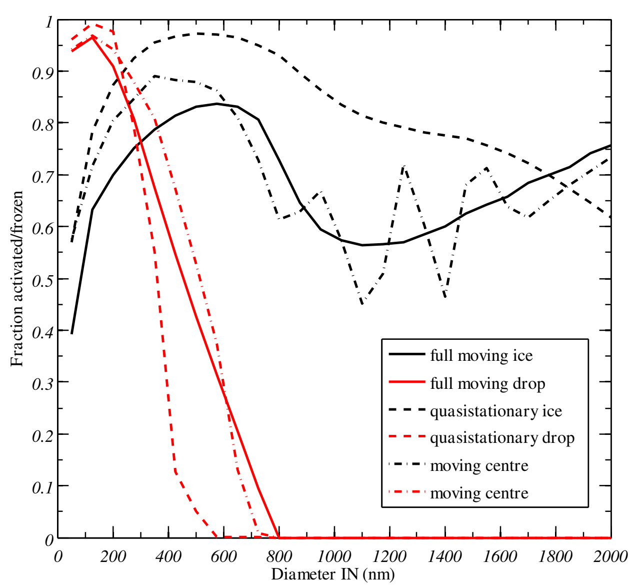

Figure S1. Comparison of the three bin structures available in ACPIM, full moving, quasistationary and moving center. For more details on these bin structures refer to Jacobson (1999). One mode of INP with a concentration of 10 cm^{-3} was used here.

2 Idealised suppression case

Supplementary Fig. S2. shows an idealised case demonstrating the suppression of ice formation caused by the presence of CCN when the criteria for heterogeneous freezing is *Activated only*.

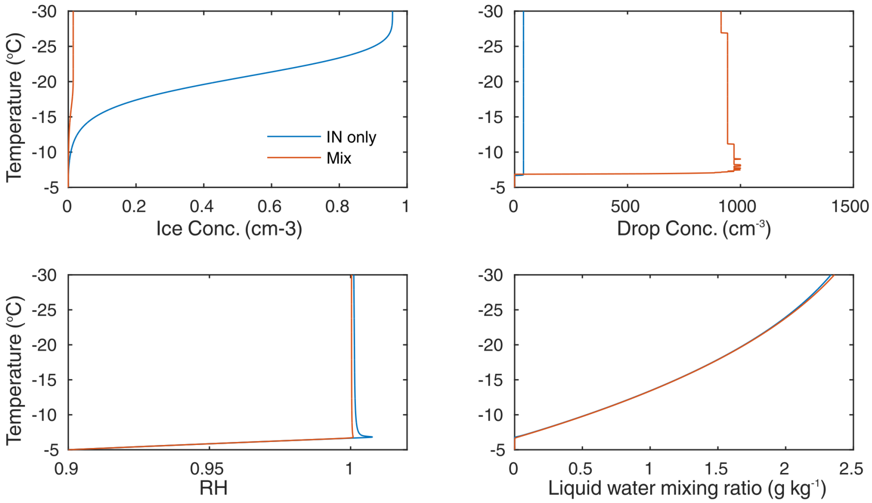

Figure S2. The *low CCN* case (blue lines) contained 1 L^{-1} 300 nm K-feldspar and 50 cm^{-3} 60 nm ammonium sulphate particles. The *high CCN* case (red lines) contained 1 L^{-1} 300 nm K-feldspar and 2000 cm^{-3} 200 nm ammonium sulphate particles. Initial conditions for the two simulations are the same as those detailed in Table 3. An updraft velocity of 0.5 ms^{-1} was used in both cases. The criteria for heterogeneous freezing was that only activated INPs could freeze. Top left panel shows the number concentration of ice with decreasing temperature. Top right panel shows the droplet number concentration with decreasing temperature. Bottom left panel shows the relative humidity with respect to water with decreasing temperature. Bottom right panel shows the liquid water mixing ratio with decreasing temperature.

3 Example lognormal fits for aerosol in chamber experiments

Supplementary Figures S3 and S4 give examples of the lognormal fits used to represent the aerosol present in the chamber in model simulations.

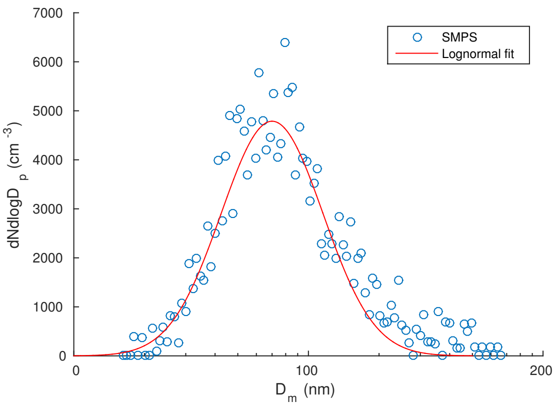

Figure S3. Example lognormal fit for ammonium sulphate particles. Blue circles are measurements made by the SMPS instrument while the red line is the lognormal fit used as initial conditions in ACPIM.

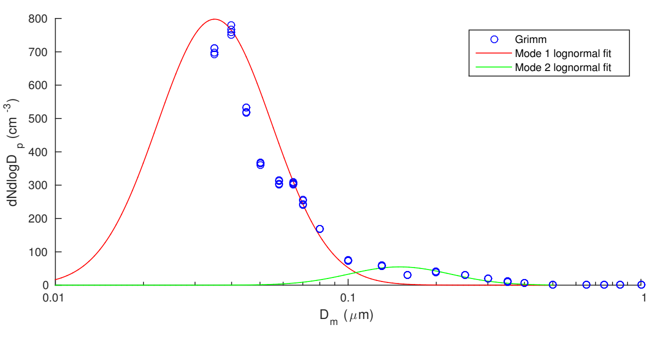

Figure S4. Example lognormal fit for K-feldspar particles. Blue circles are measurements made by the GRIMM instrument while the red line is the lognormal fit used as initial conditions in ACPIM.

4 Fit used for freezing temperature of solution

Supplementary Figure S5 shows the fit used here for the freezing temperature of a solution when $logJ$ =0 $\mathrm{cm^{-3}s^{-1}}$ following the parameterisation for homogeneous freezing by Koop et al. (2000).

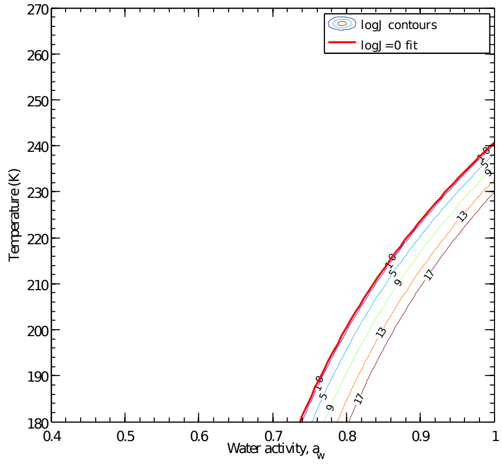

Figure S5. Contours of the homogeneous freezing, J, rate calculated according to the Koop parameterisation, (Koop et al., 2000). Red line is the polynomial fit to the *logJ=0* contour.

5 Additional sensitivities of the suppression effect results

The following figure is similar to Fig. 5. and Fig. 7. however the criteria for freezing is that there must be sufficient water on an INP to overcome the freezing point depression caused by the presence of solutes.

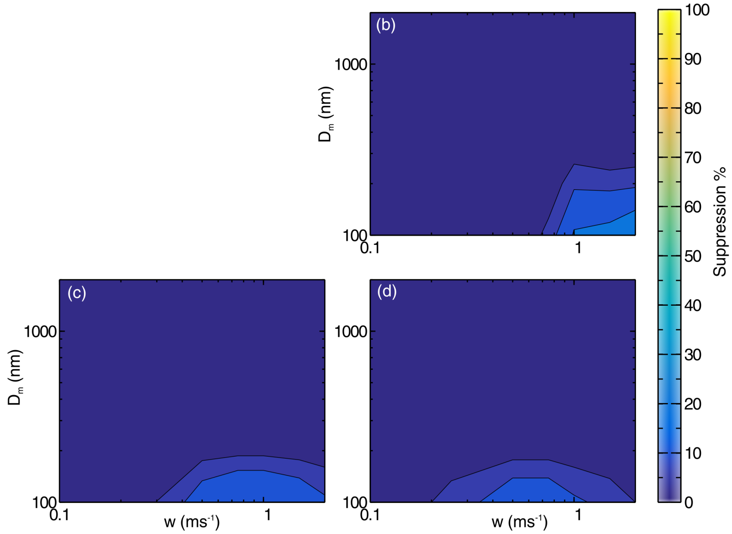

Figure S6. Results from 147 pairs of *low CCN* and *high CCN* model simulations with initial conditions as detailed in tables 3 and 4. the ice crystal number concentration in every simulation was taken at -30^{o}C. Contours show the percentage less ice that formed in the *high CCN* case compared to the *low CCN* case. The criteria for freezing in these results was that there was sufficient water mass on an INP to overcome the freezing point depression caused by the presence of solutes.

The following six figures display results from the same model runs as those shown in Fig. 5., Fig. 6. and Fig. 7. however compare ice crystal number concentrations at temperatures warmer than -30^{o}C. The criteria for freezing in Supplementary Fig. S7. and S8. is *Activated only*. The white areas on the top right panel of Supplementary Fig. S7. and the top two panels Supplementary Fig. S8 show negative suppression. This means more ice had formed in the *high CCN* case compared to the *low CCN* case at the point the comparison between the two cases was made. At most this is -2.4 %. At the end of the simulation time, around -30^{o}C shown in Figure 5, there is always higher, or the same, number concentration of ice in the *low CCN* case.

The criteria for freezing in Supplementary Fig. S9. and S10. is A_w and M_{cw} in Supplementary Fig. S11. and S12.

Additional sensitivity simulations with a representation of desert dust, following Niemand et al. (2012), instead of K-feldspar were preformed. The results from these simulations with three different freezing criteria (*Activated Only*, A_w and M_{cw}) are presented in the Supplementary Figs. S13, S14 and S15.

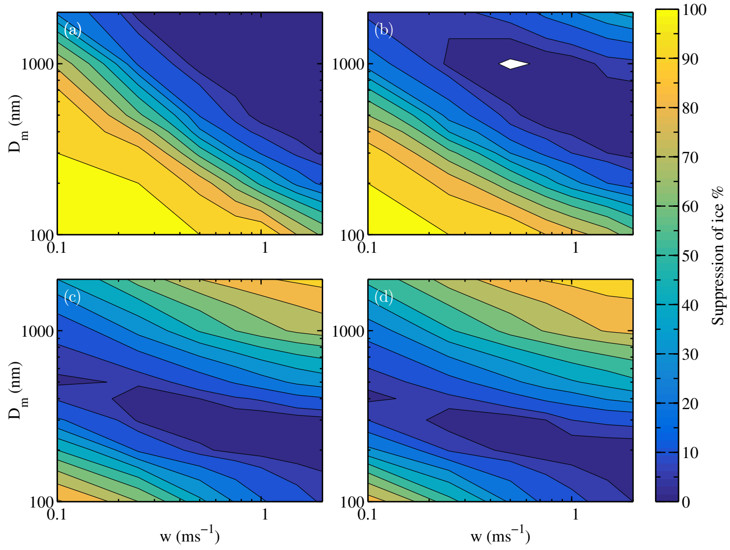

Figure S7. Same model runs as displayed in Figure 5 however with ice concentrations compared at -15^{o}C instead of -30^{o}C.

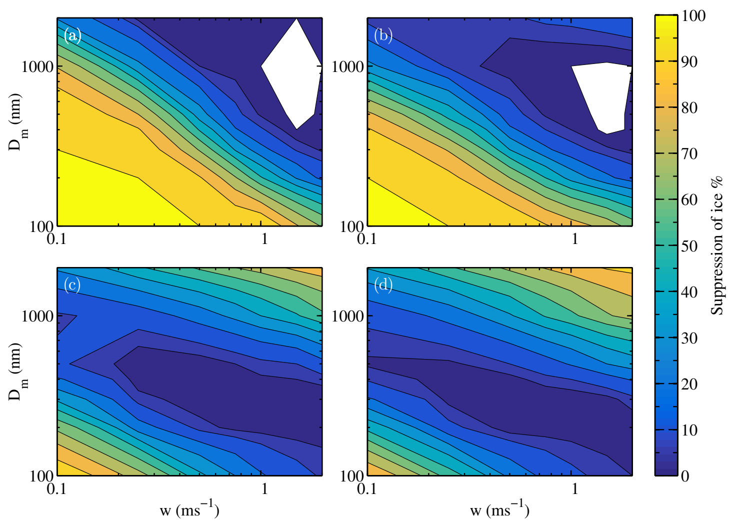

Figure S8. Same model runs as displayed in Figure 5 however with ice concentrations compared at -20^{o}C instead of -30^{o}C.

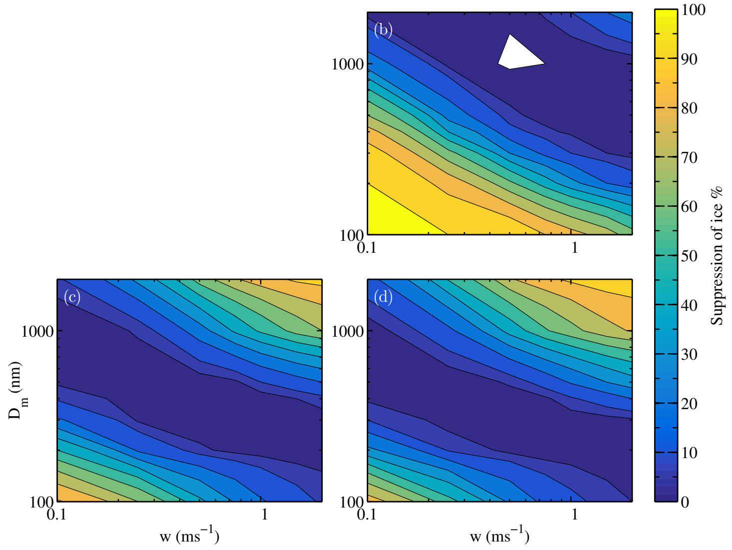

Figure S9. Same model runs as displayed in Figure 6 however with ice concentrations compared at -20^{o}C instead of -30^{o}C.

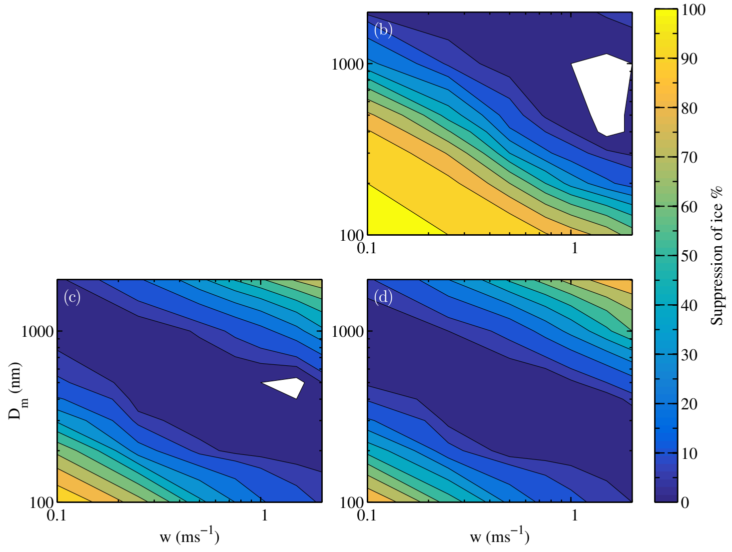

Figure S10. Same model runs as displayed in Figure 6 however with ice concentrations compared at -20^{o}C instead of -30^{o}C.

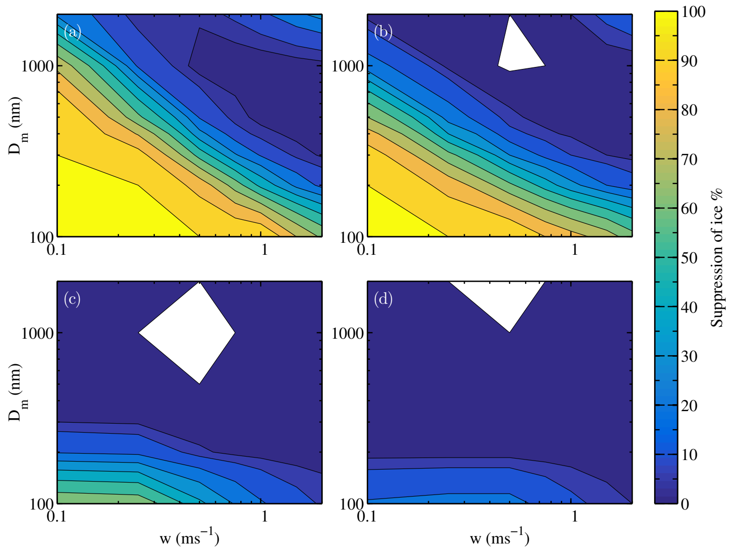

Figure S11. Same model runs as displayed in Figure 7 however with ice concentrations compared at -20^{o}C instead of -30^{o}C.

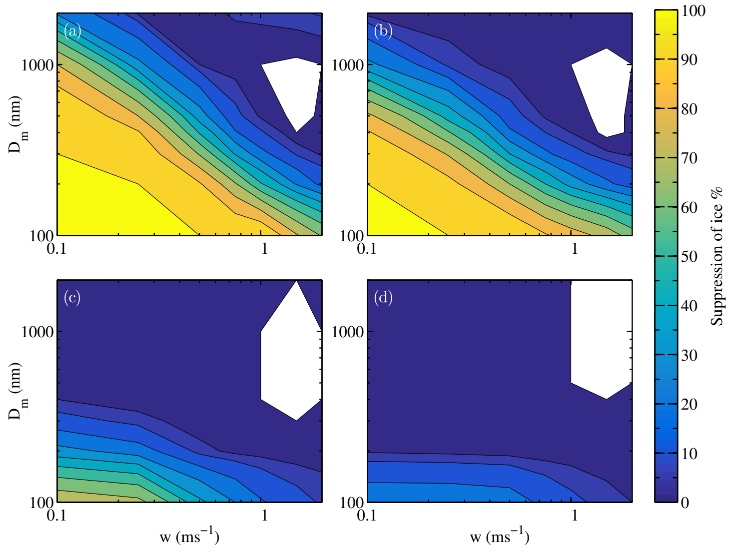

Figure S12. Same model runs as displayed in Figure 7 however with ice concentrations compared at -20^{o}C instead of -30^{o}C.

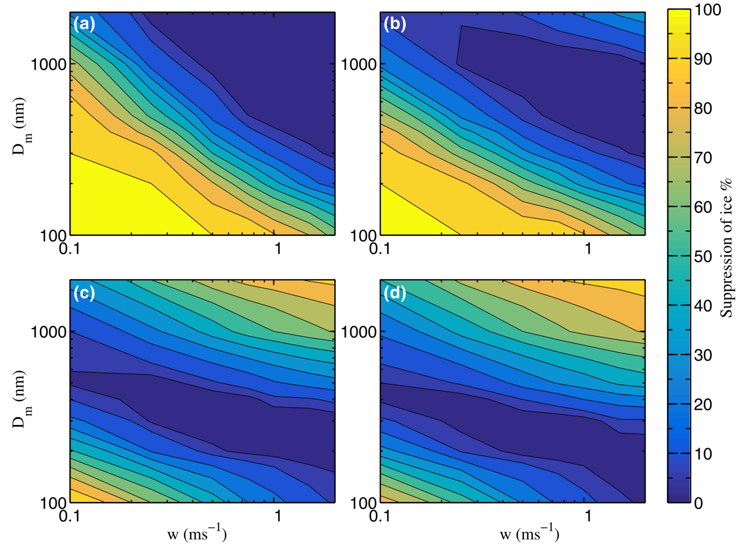

Figure S13. Results from 196 pairs of *low CCN* and *high CCN* model simulations with initial conditions as detailed in Tables 3 and 4 however instead of K-feldspar a representation of desert dust is used for INPs. For each of the four soluble fractions of INP the updraft velocity ranged between 0.1 and 2 ms^{-1} and the diameter of the INP varied between 100 and 2000 nm. The ice crystal number concentration in every simulations was taken at -30^{o}C. Contours show the percentage less ice that formed in the *high CCN* case compared to the *low CCN* case. The criteria for heterogeneous freezing in these results is only activated drops can freeze.

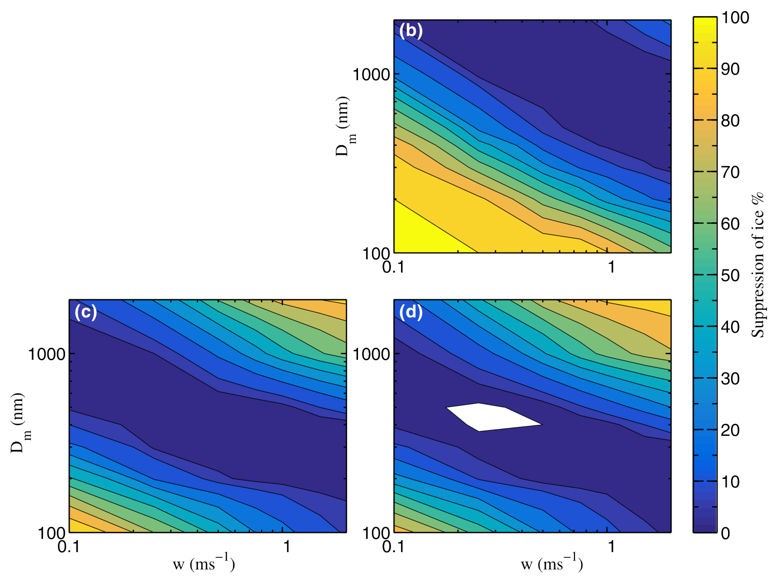

Figure S14. Same as Supplementary Figure S13 however the criteria for freezing is A_w.

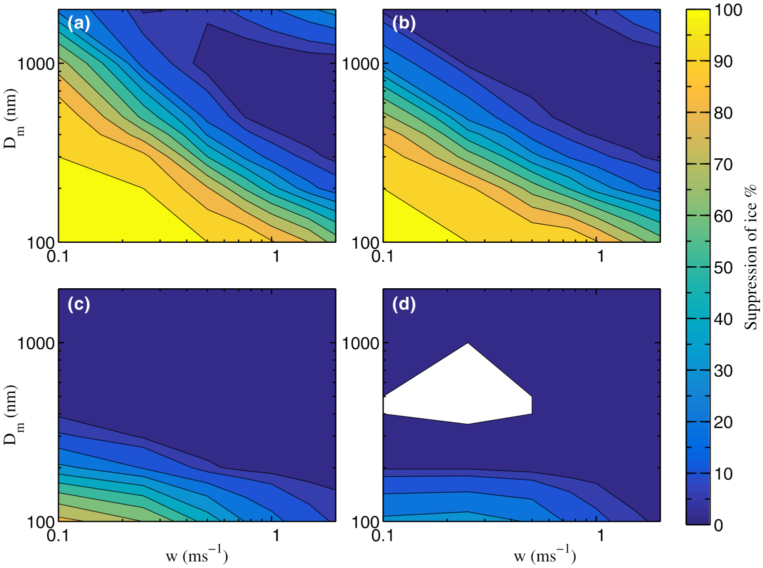

Figure S15. Same as Supplementary Figures S13 and S14 however the criteria for freezing is M_{cw}.

6 Comparison of the mass of water required for freezing for three freezing criteria

Under the chamber conditions in this study, T = -25^{o}C, a smaller mass of water is required for a particle with a diameter $<$ 0.8 μm that is 1% soluble to activated than is required to reach M_{cw} or A_w. Supplementary Fig. S16a shows the mass of water required by each of the three criteria for freezing, M_{cw}, A_w and *Activated Only*, for different particle diameters under chamber conditions. The mass of water required to reach M_{cw} and A_w is similar for particles with diameters $<$ 0.5 μm. At higher temperatures and for particles with larger soluble fractions the mass of water required to activate a particle is greater than the mass required to reach M_{cw} or A_w, this difference is greatest at $D_p > 1$ μm. This is shown in Supplementary Fig. S16b which is similar to Supplementary Fig. S16a, however T = -7^{o}C in the calculation of mass of water required for activation and particles are 25% soluble. Under the conditions in Fig. S3b, and for particles with Dp $>$ 1.5 μm, the mass of water required to reach activation is greater than M_{cw} and A_w. This means that a particle which does not have enough condensed water to activate and therefore cannot freeze under the criteria of *Activated Only*, could freeze if the criteria for freezing was either M_{cw} or A_w where a smaller mass of water is required for freezing.

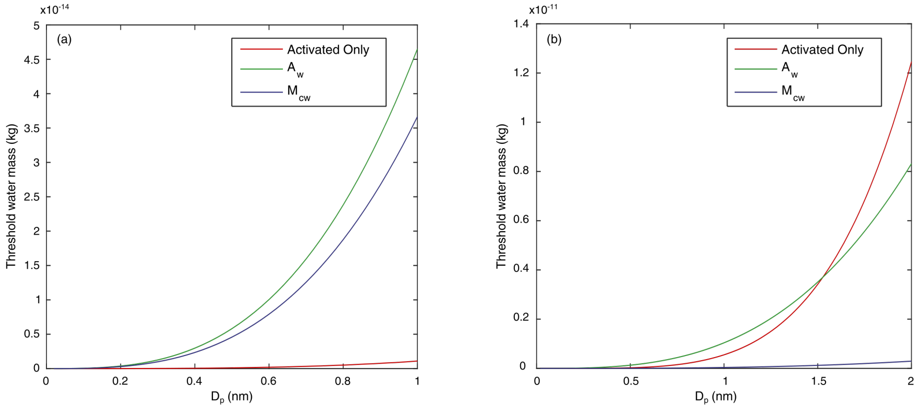

Figure S16. a) A comparison of the threshold mass of water required for freezing for three difference criteria for different particle diameters under chamber conditions. b) Similar as Fig. S3a however for 25% soluble dust particles under similar conditions as found in parcel model simulations, the results of which are shown in Fig 3a-c.

References

Jacobson, M. Z.: “Fundamentals of atmospheric modelling”, “Cambridge University Press”, 1999.

Koop, T., Luo, B., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature, 406, 611–614, 2000.

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., and Leisner: A particle-surface-area parameterisation of immersion freezing on desert dust particles, Journal of Atmospheric Sciences, 69, 3077–3092, 2012.