Supplementary Material for the ACP Manuscript, Heterogeneous freezing in synoptic cirrus enables subsequent homogeneous freezing

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This supplementary material provides additional information on ice nucleation parameterisation schemes, supporting figures and some additional sensitivity test results.

1 Sensitivity of wind fluctiations to changes in domain width and grid resolution

A sensitivity test was conducted to look into the sensitivity of the wind fluctuations by changing the horizontal domain horizontal width from 3 km to 6 km. Also, a simulation run with lower resolution Δx$=150$ m and 18 x 18 km domain was conducted. Figure S1 shows vertical wind at approximately 11 km altitude. The 18 km domain resolved large wavelengths while there was a lack of shorter wavelengths resolved. The 6 km domain resolved longer wavelengths as well as shorter wavelengths. This is due to the higher resolution of Δx$=50$ m. The 3 km model domain resolved only small scale waves. Also, the horizontal variability of temperature and humidity did not differ significantly. In contrast, the 18 km domain had no significant horizontal variations in temperature and humidity due to unresolved small scale waves. Based on these results, the 3D simulations were conducted using a 3 km domain, as it provided wind variability comparable to that of larger domains while significantly minimizing computational costs.

On the other hand, the bottom right plot in Figure S1 shows the vertical wind when the ice nucleation scheme was turned off. It is clear that the ice nucleation had no major correlation with the vertical wind fluctuations.

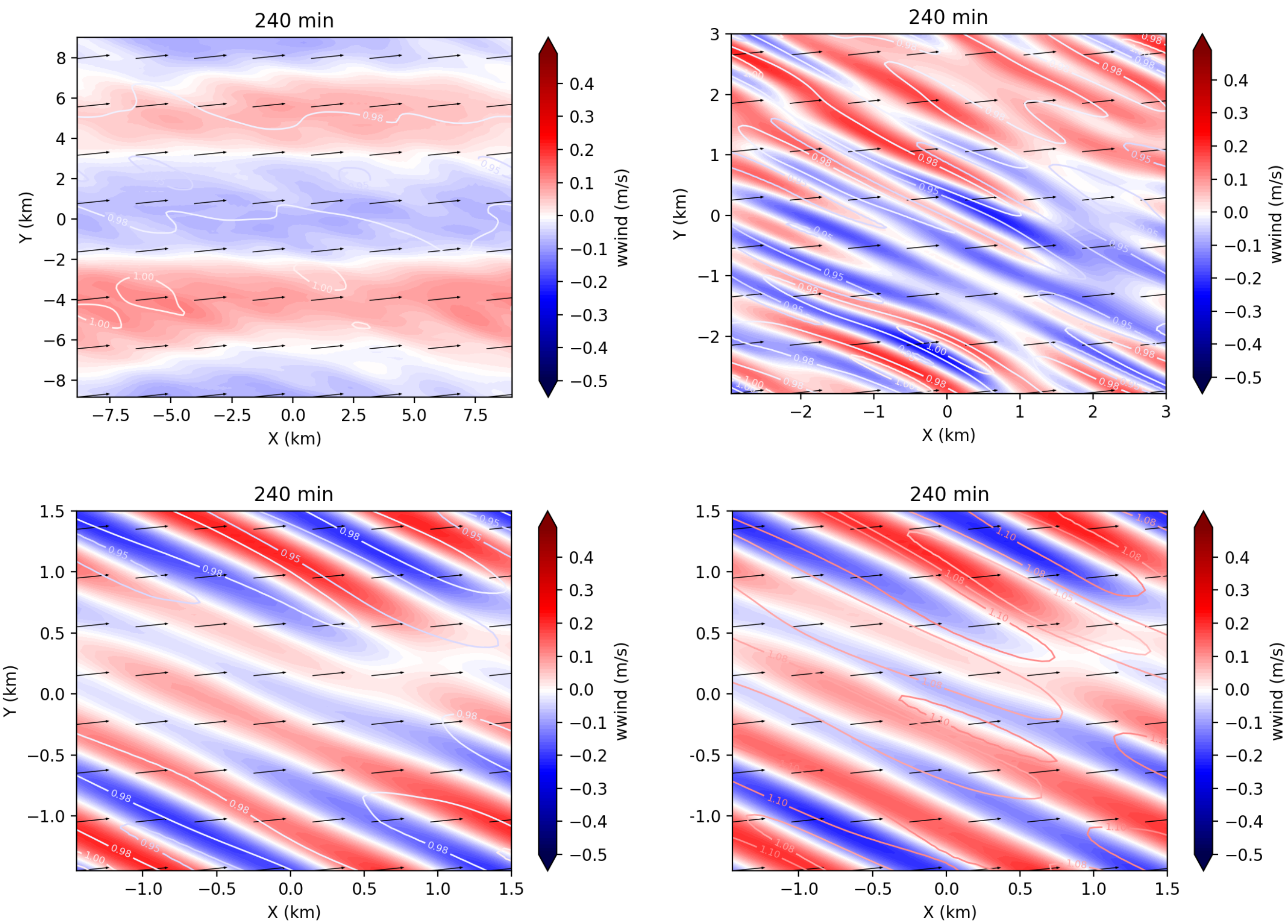

Figure S1. Vertical wind in 18 km (upper left), 6 km (upper right) and 3 km domain (lower left) at 3 hours into the simulation on 11 km altitude. Lower right plot shows the case without ice nucleation. S_i is indicated with colored contours. The vectors represent the horizontal wind direction and magnitude.

2 Supplementary figures

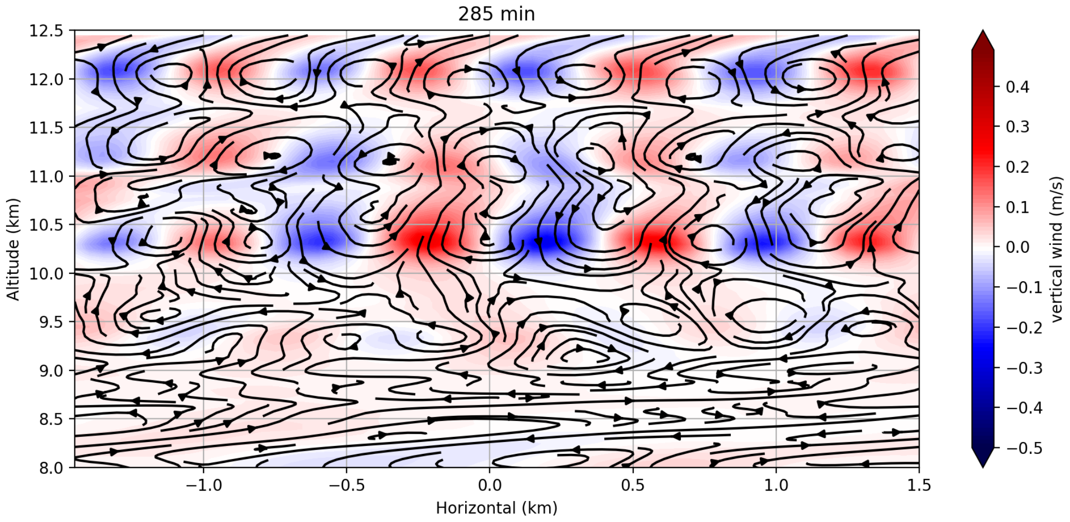

Figure S2. Gravity waves induced by vertical wind shear. Horizontal eddies shown with streamlines on y-axis plane.

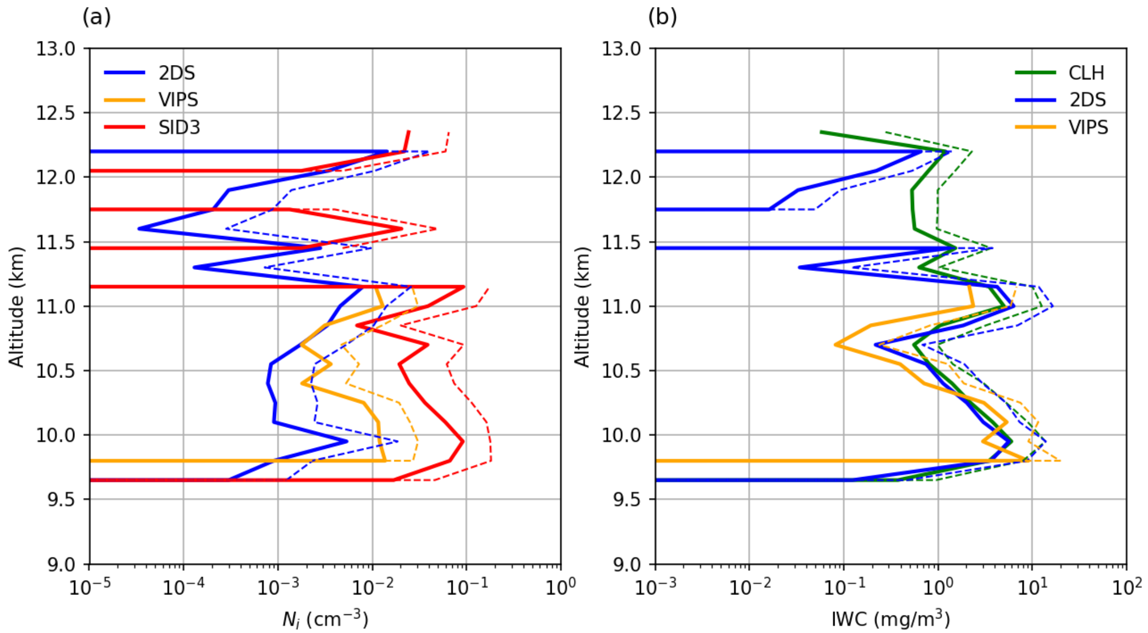

Figure S3. Observed vertical structure of N_i during the MACPEX mission flight. The data is binned into Δz=150 m wide bins. The dashed lines represent 1-sigma standard deviation of a bin.

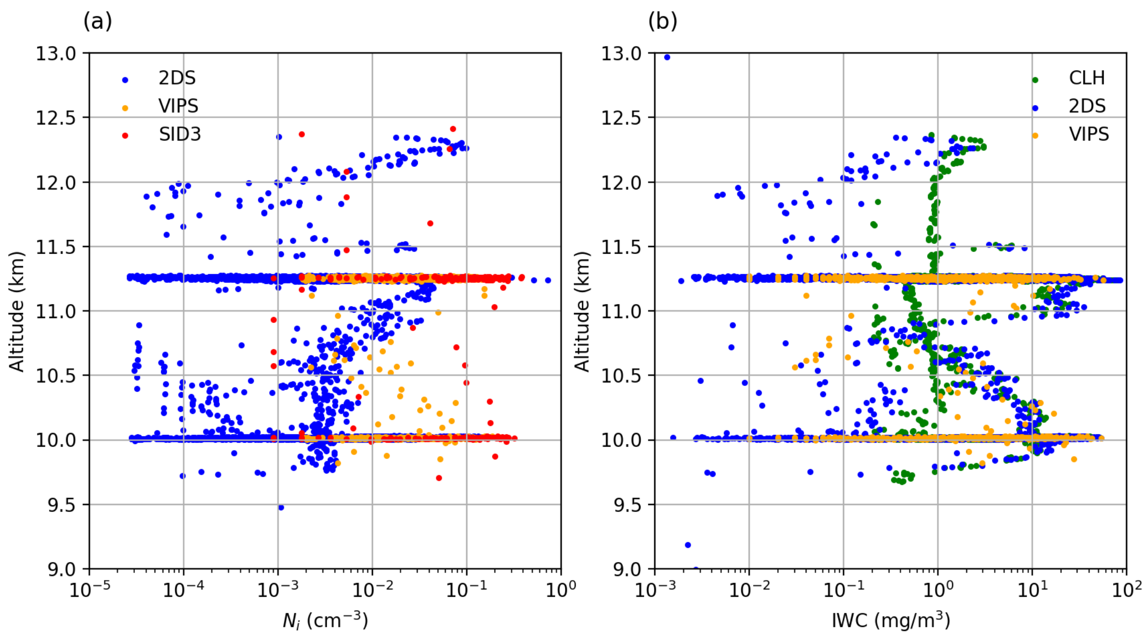

Figure S4. Observed values N_i during the MACPEX mission flight for the instruments that measured N_i.

3 Description of ice nucleation parameterization scheme by Ullrich et al. (2017)

Ullrich et al. (2017) parameterization scheme for deposition ice nucleation, is based on 11 years worth of laboratory experiments conducted in the Aerosol Interaction and Dynamics in the Atmosphere (AIDA) chamber. This framework was specifically developed to assess the nucleation characteristics of mineral dust and soot particles under conditions relevant to cirrus clouds (Ullrich et al., 2019). The parameterization presented below quantifies the nucleation ability of mineral dust/soot particles in terms of ice nucleation active site (INAS) density n_s (e.g., Hoose and Möhler, 2012; Vali, 2015). The INAS density represents the number of active sites per unit surface area of an INP (m^{-2}) at a specific temperature and S_i. These active sites are locations on the particle surface where nucleation occurs more effectively, typically resulting from surface inhomogeneities (Vali et al., 2015). The parametrization is given by:

$$n_s = \exp\{\alpha(S_i - 1)^{1/4}\cos[\beta(T-\gamma)]^2 \mathrm{arccot}[\kappa(T-\lambda)]/\pi\}$$
$$\mathrm{arccot(x)} = \frac{\pi}{2} - \mathrm{arctan(x)} \tag{1}$$

where the parameters and corresponding values for mineral dust are given in Table 1.

	α	β	γ	κ	λ	Valid temperature range (K)
Mineral dust	285.692	0.017	256.692	0.080	200.745	[206, 240]

Table 1. Fit parameters for equation 2.11 for mineral dust particles. The parametrization is valid for S_i below water saturation, homogeneous nucleation threshold and above 1.0.

Predicted frozen fraction FF of INPs can be calculated by multiplying the INAS density by the surface area of INPs with a single particle size.

$$FF = n_s S_X = n_s \pi D_X^2 \tag{2}$$

4 Homogeneous freezing parametrization by Koop et al. (2000)

The parametrization for homogeneous freezing follows Koop et al. (2000) formulation. The nucleation rate is parametrized with shift in melting curve $\Delta a_w = a_w - a_w^i$. In Koop (2015), the $a_w^i = \frac{p_{\mathrm{ice}}(T)}{p_{\mathrm{liq}}(T)}$. Knowing this an equation follows:

$$\Delta a_w = a_w - a_w^i = \frac{p_v}{p_{\mathrm{liq}}(T)} - \frac{p_{\mathrm{ice}}(T)}{p_{\mathrm{liq}}(T)} \tag{3}$$

Koop et al. (2000) parametrizations produces freezing rate ($cm^{-3}s^{-1}$) as:

$$\log(J) = -906.7 + 8502\Delta a_w - 26924(\Delta a_w)^2 + 29180(\Delta a_w)^3 \tag{4}$$

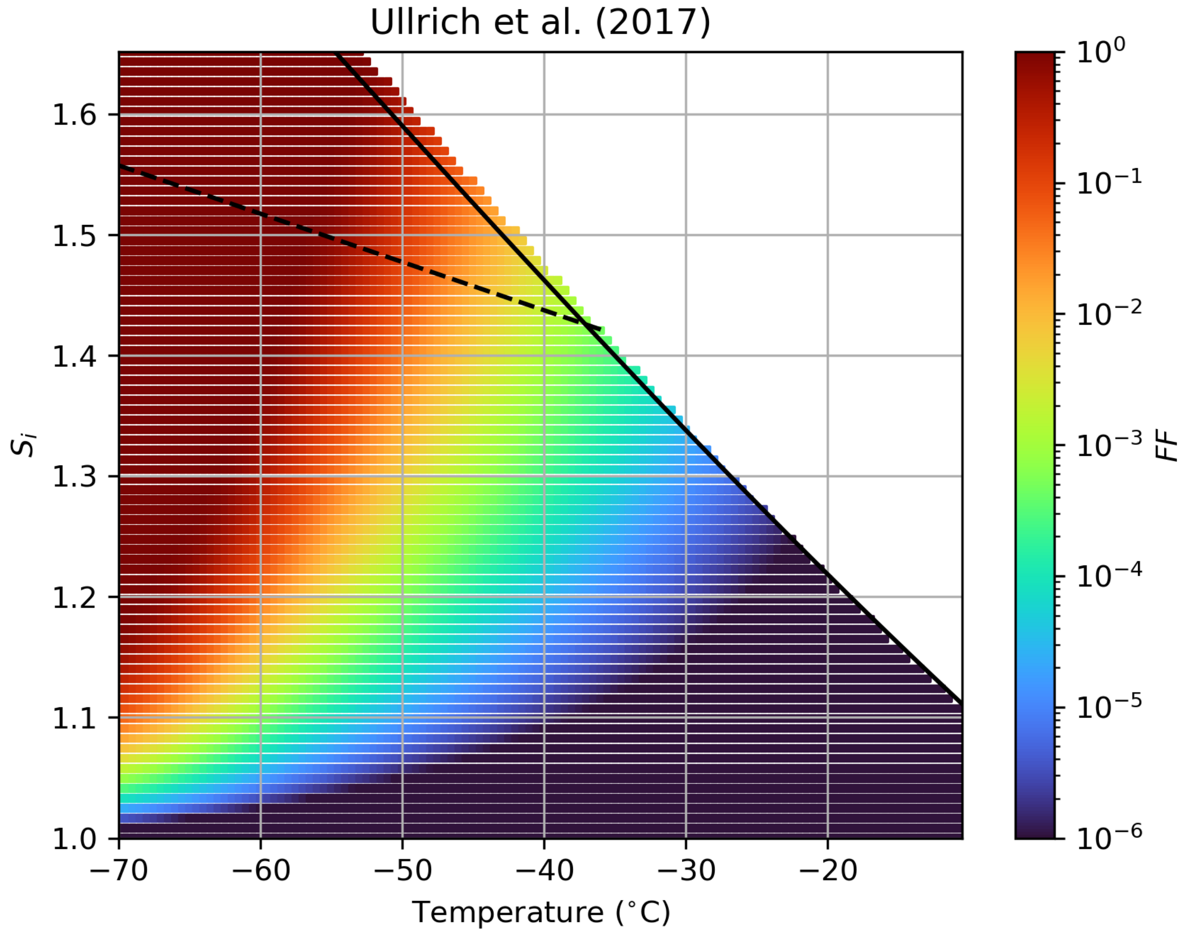

Figure S5. Frozen fraction of mineral dust particles used in this study. The activity is integrated over the mineral dust particle sizes to reflect the total activity. Dashed line indicates Koop et al. (2000) line at the critical threshold for homogeneous freezing. Solid line follows water saturation (Murphy and Koop, 2005).

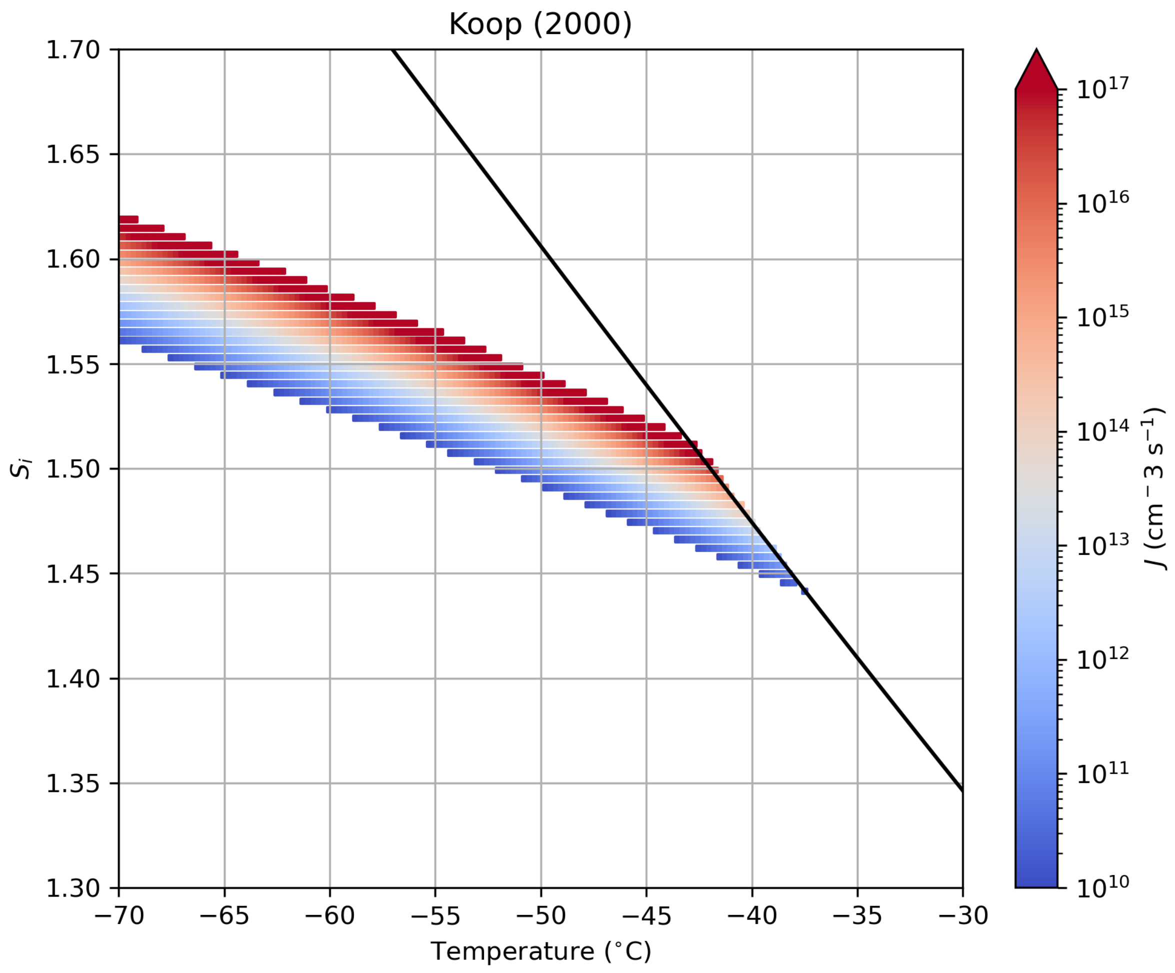

Figure S6. Koop et al. (2000) parametrization scheme as a function of temperature and Δa_w. Solid line indicates the water saturation line.

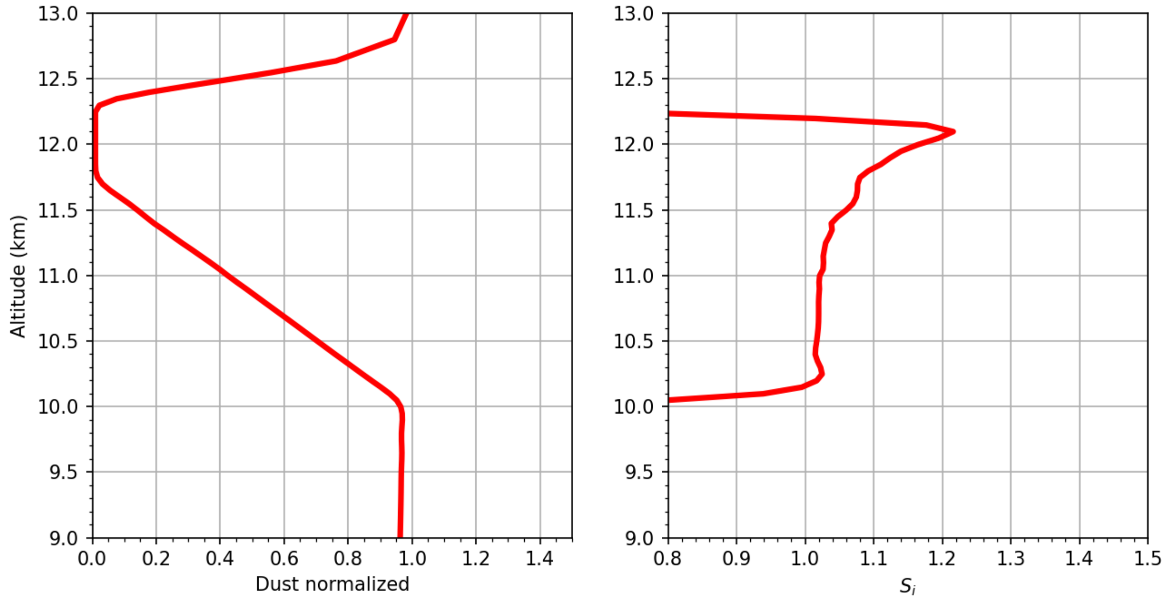

Figure S7. Initial profile of dust and S_i for adjusted profile runs.

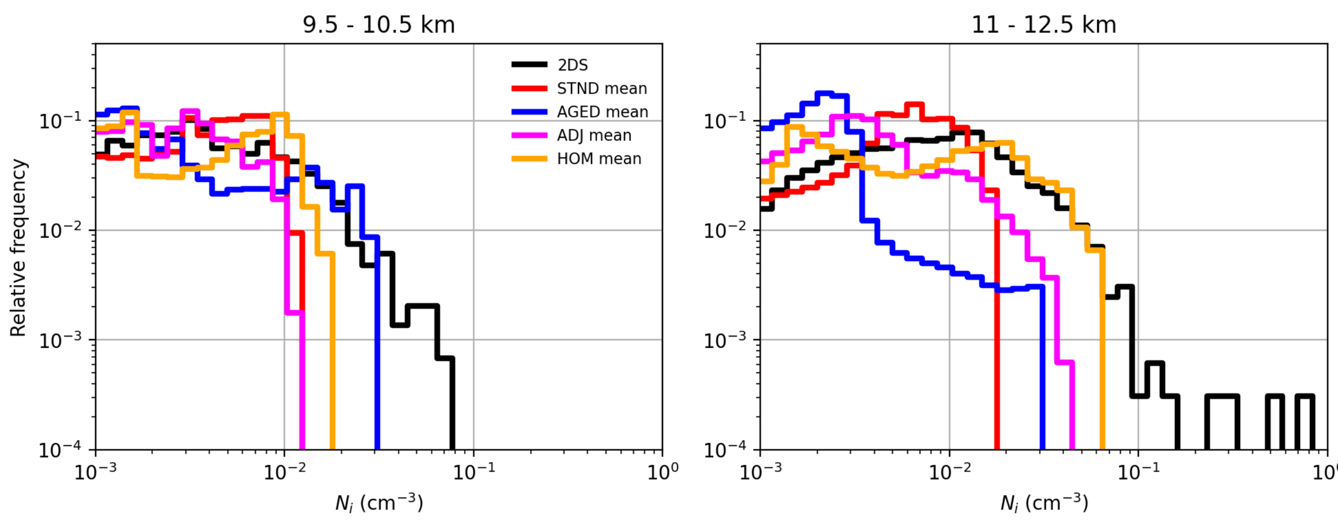

Figure S8. Frequency distributions for all setups in the study with 1D-column setup.

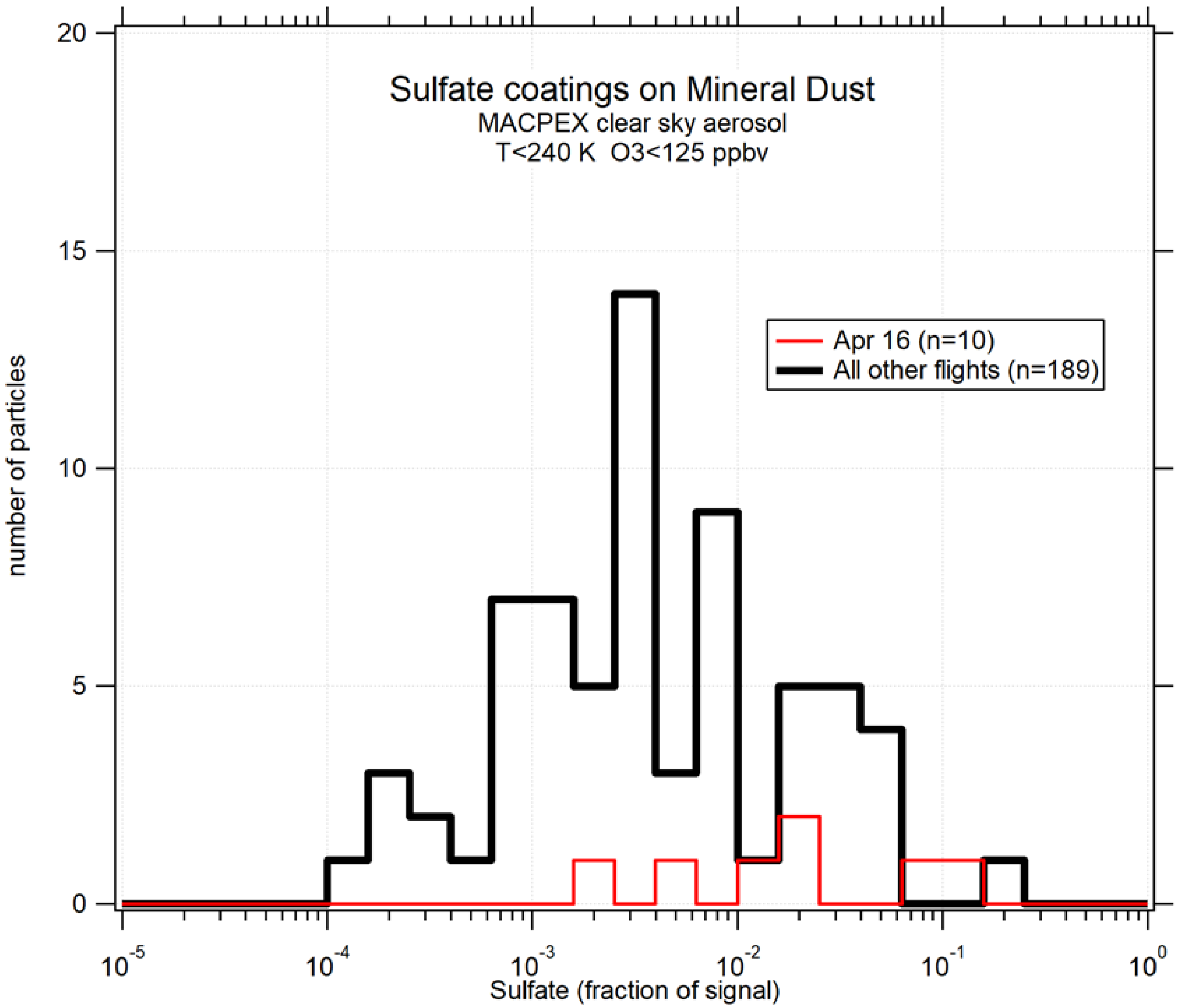

Figure S9. The raw sulfate signal on all clear-sky mineral dust spectra, comparing Apr 16 cirrus regime with all other flight days. The statistics are poor, but this "is consistent with" the idea that well-aged UT air gives rise to thicker coatings on dust particles that could potentially reduce their heterogeneous nucleation ability.

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