Supplement of Atmos. Meas. Tech., 18, 5223–5245, 2025
https://doi.org/10.5194/amt-18-5223-2025-supplement

Atmospheric Measurement Techniques
Open Access
EGU

Supplement of

Analyzing the chemical composition, morphology, and size of ice-nucleating particles by coupling a scanning electron microscope to an offline diffusion chamber

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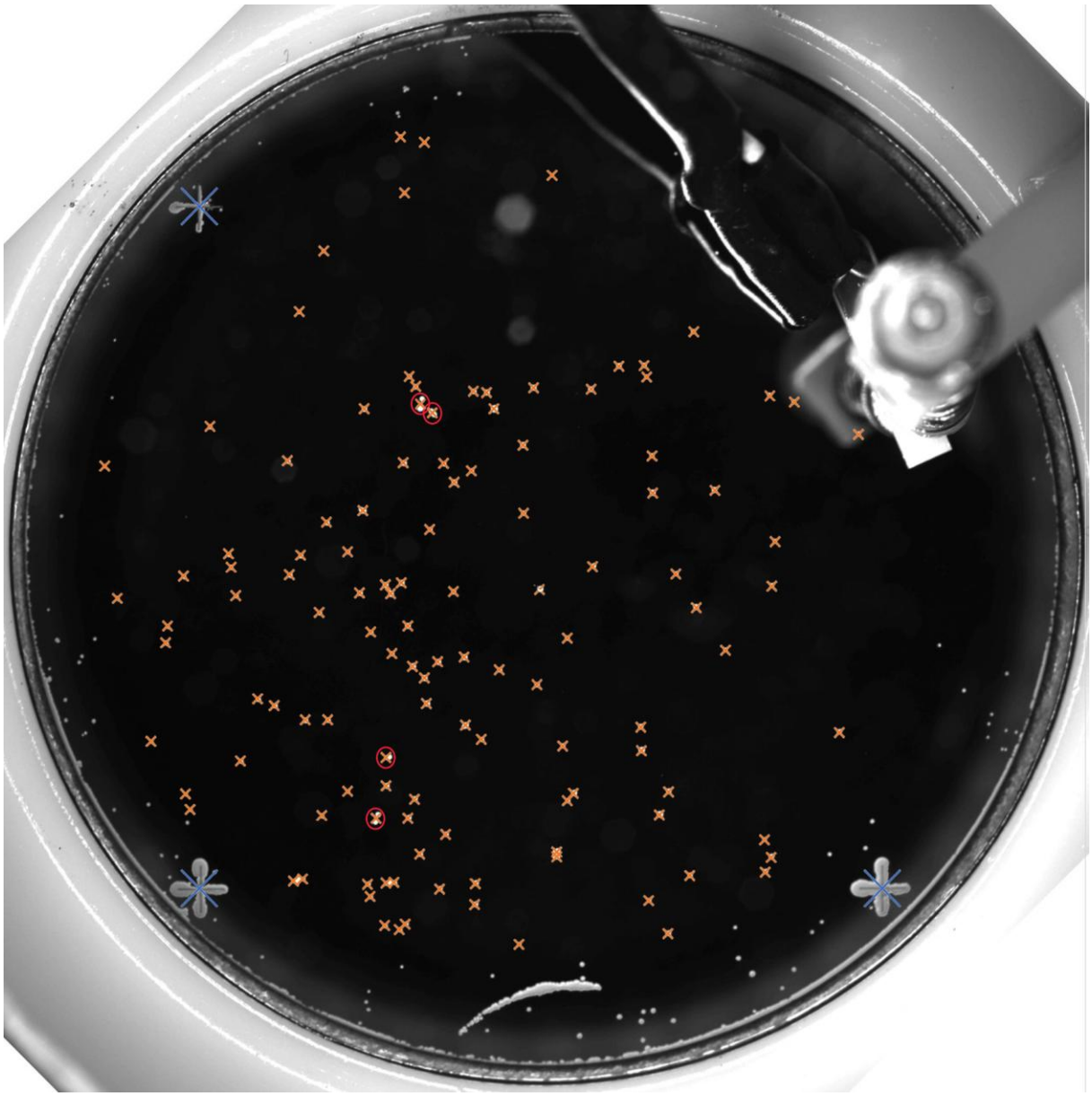
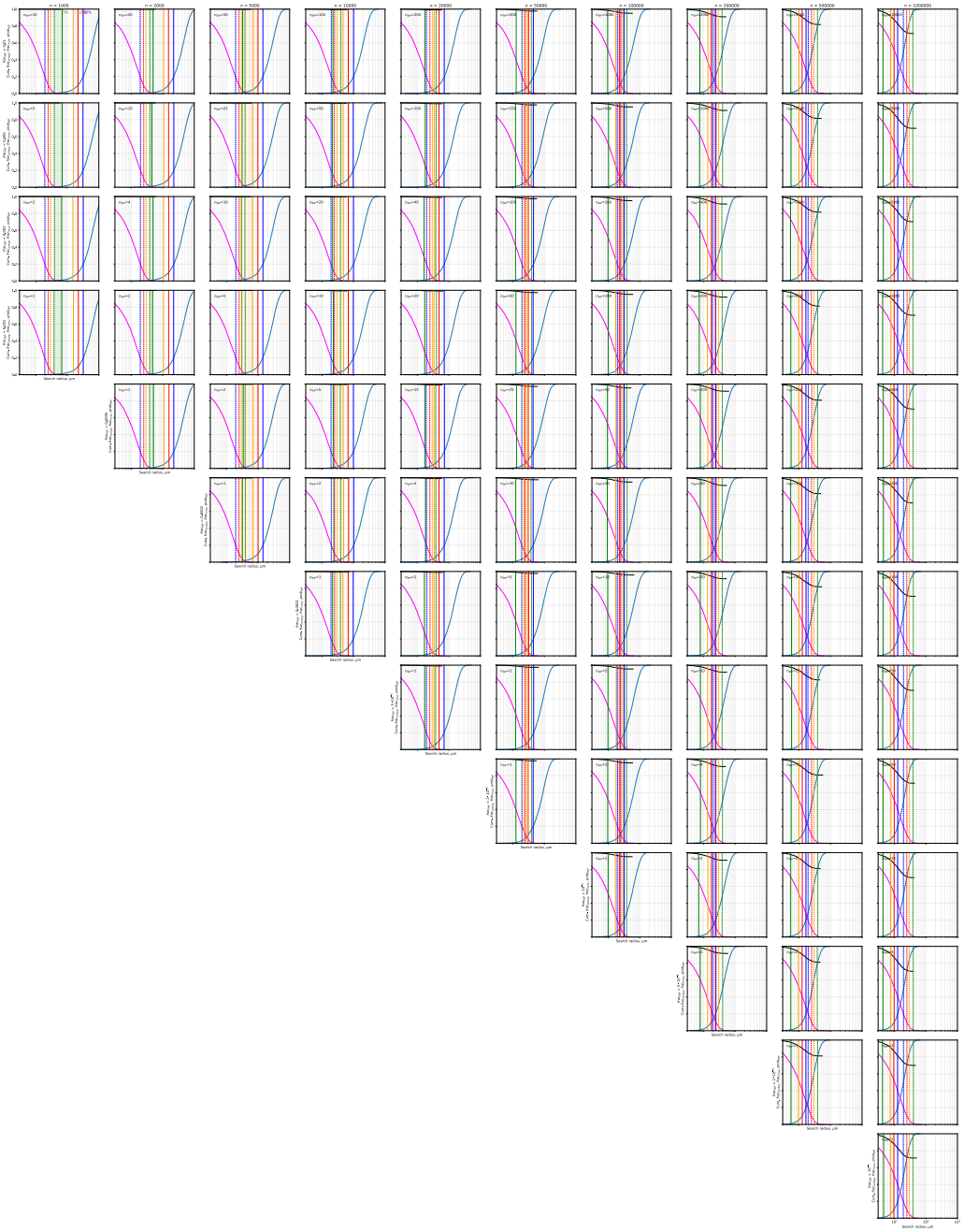
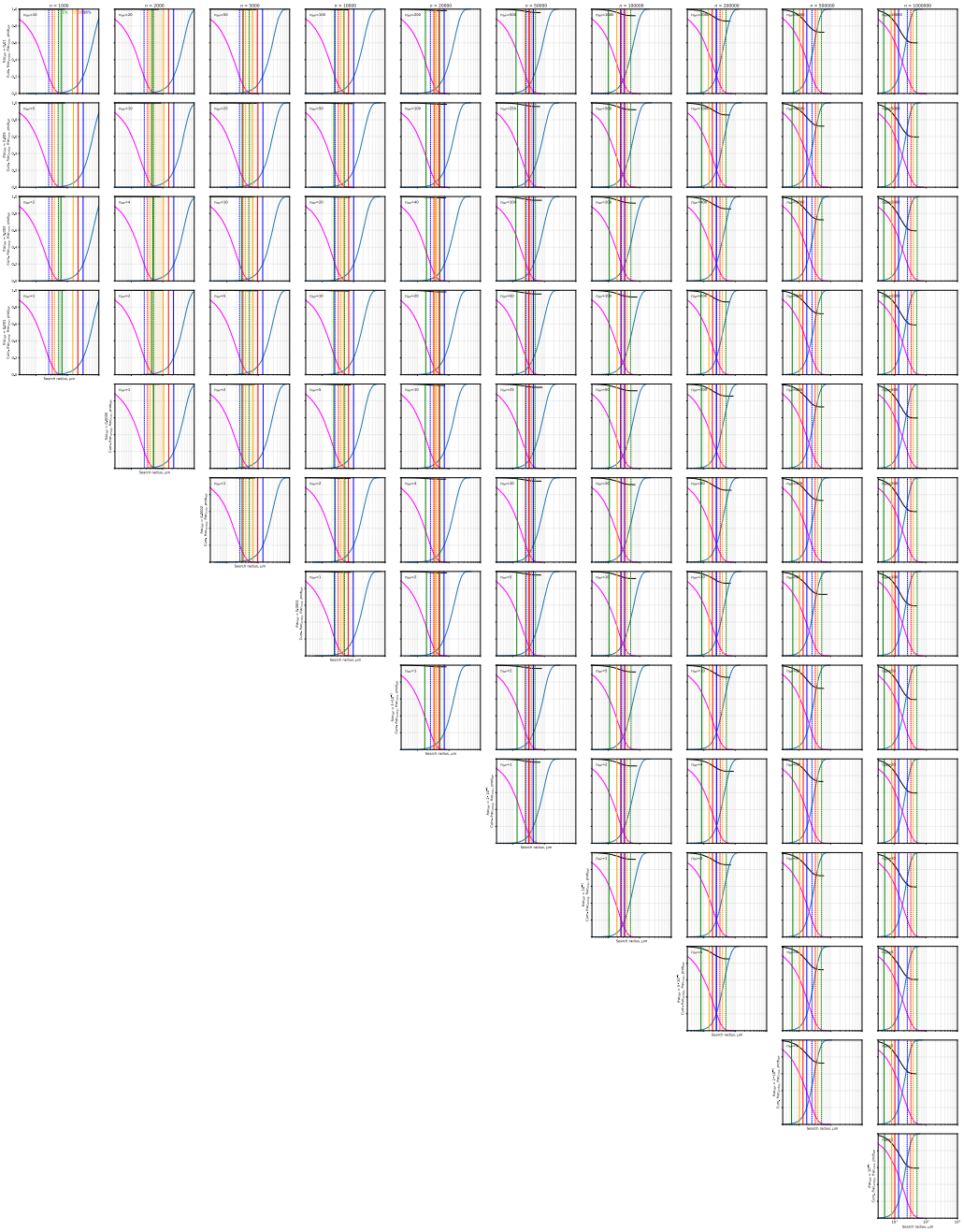


Figure S1: Image of a silicon wafer from the FRIDGE diffusion chamber measurement with grown ice crystals (white dots) and the corresponding calculated positions for SEM (orange crosses). Miscalculated coordinates are marked with a red circle. The three laser-engraved crosses (large blue crosses) represent the reference points for the coordinate system. The edge of the sample substrate as well as the area around the temperature sensor are excluded from analysis.

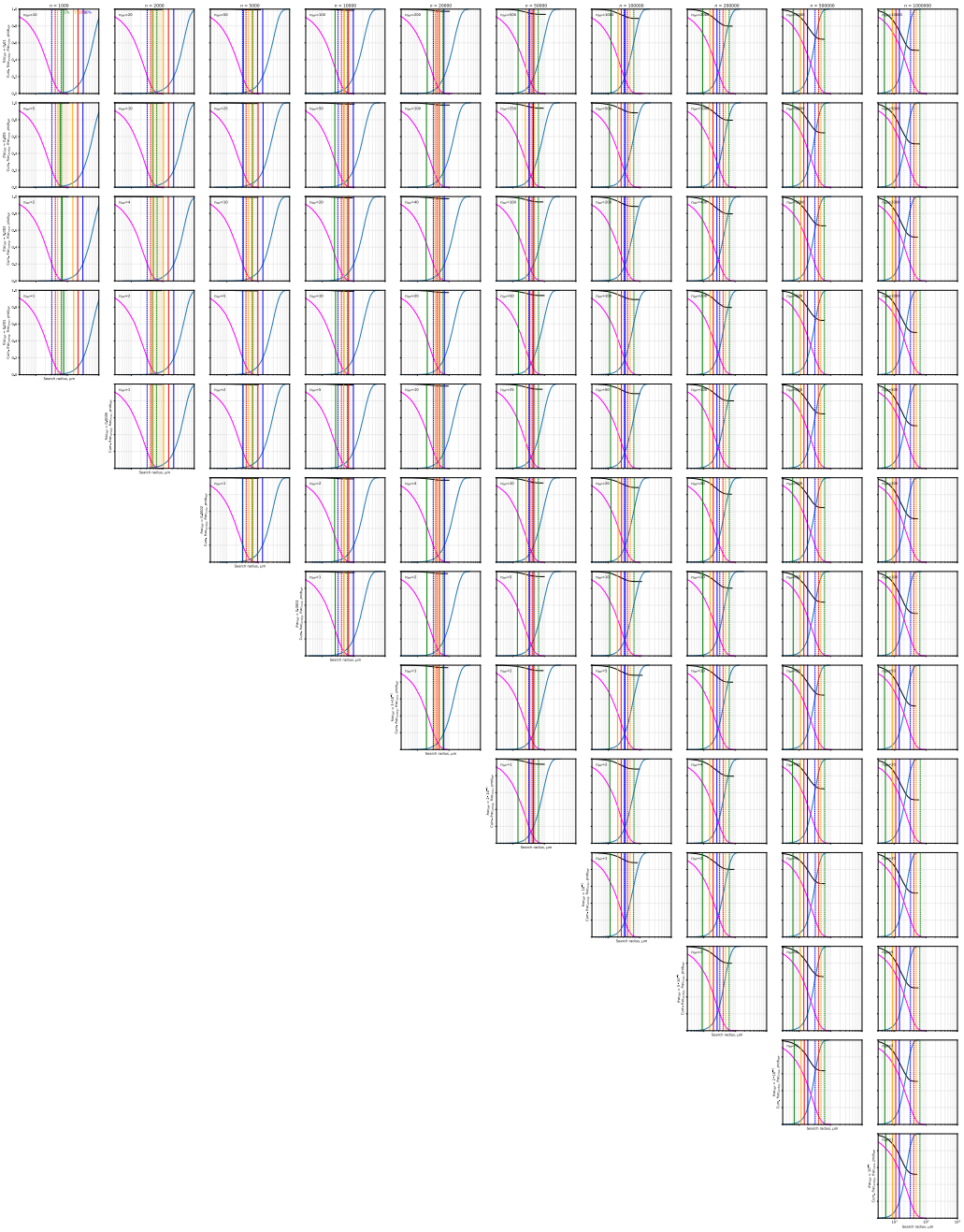
a)



b)



c)



d)

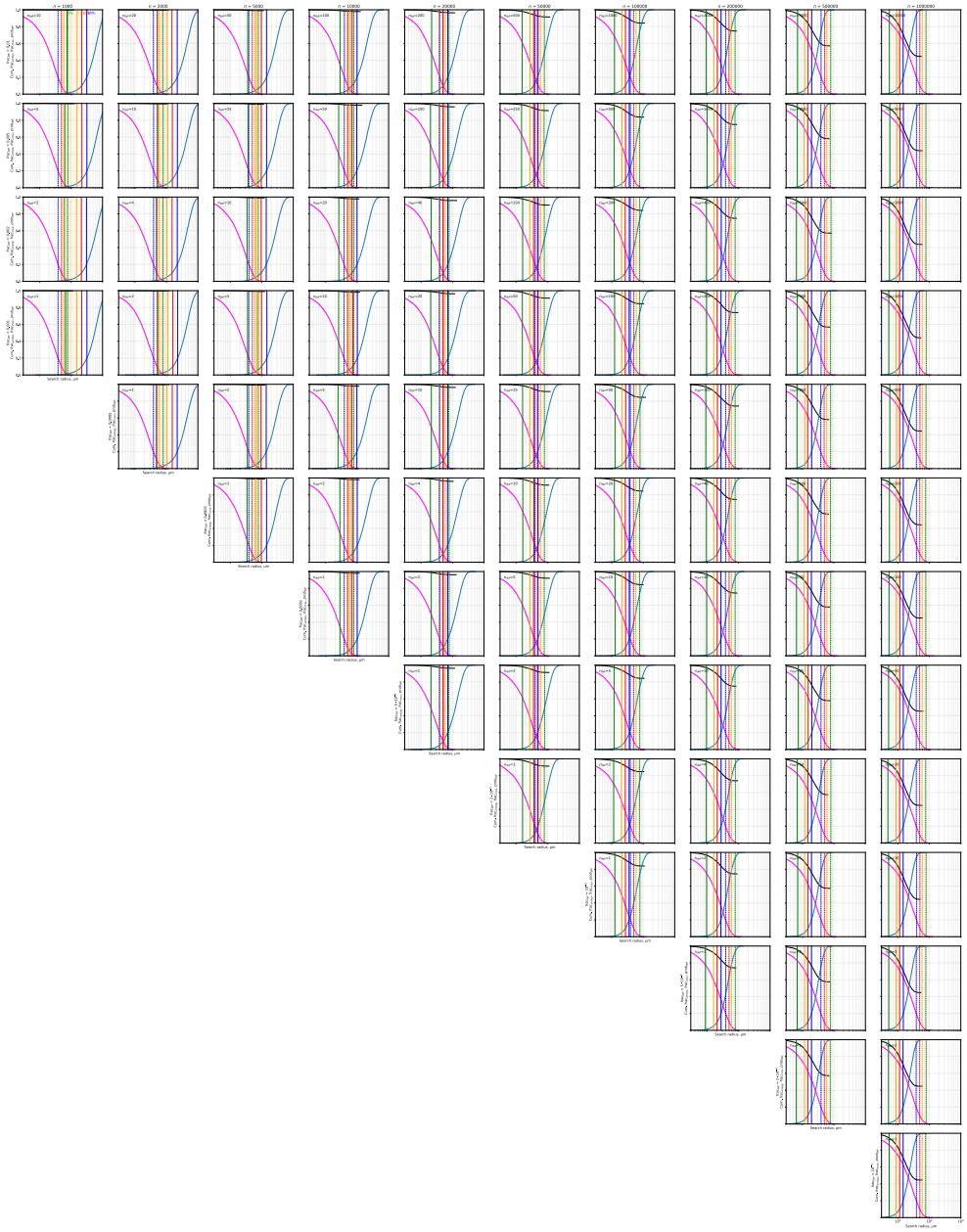


Figure S2: Model simulation results. Cumulative fraction of unrecovered particles $frac_{miss}$ (pink line), fraction of ambiguous locations $frac_{ambig}$ (blue), and fraction of correctly identified INP $prob_{INP}$ (thin black) as function of search radius for different INP fraction $frac_{INP}$ and total particle numbers n. For this plot, a standard deviation of the positioning uncertainty of a) 15 µm, b) 20µ, c) 25 µm and d) 30 µm is used. The total number of INP n_{INP} is shown for each plot. The search radius where 20% (blue), 10% (red), 5% (orange) and 1% (green) fraction / probability is reached is marked by vertical lines for $frac_{miss}$ (broken line) and for $frac_{ambig}$ (solid line). The lowest possible error probability range is indicated by a shaded area for below 20% (blue), below 10% (red), below 5% (orange) and below 1% (green). If no shaded area is visible, the error probability is greater than 20%.

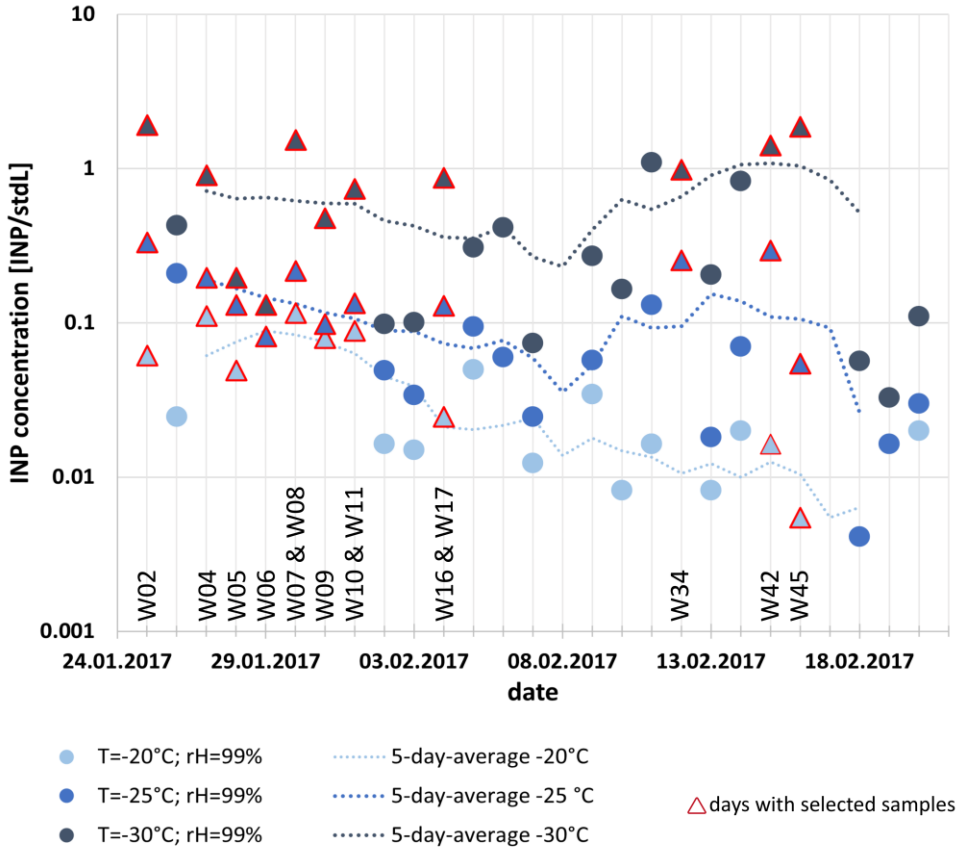


Figure S3: INP concentrations (deposition nucleation / condensation mode freezing) calculated from the FRIDGE measurements at RH = 99 %. The concentration for each sample is calculated on the basis of one measurement. For days with more than one sample, an average value was calculated. Days with analyzed samples are indicated by triangles and the corresponding sample numbers. The 5-day running average concentration is shown by the dotted lines (the figure is adapted from Weber (2019)). The error of the concentrations given here is 20% approx., based on the relative error of the counting uncertainty for individual measurements from Schrod et al. (2016).

Table S1: 0.95 confidence intervals for the chemical composition shown in Fig. 11, calculated by bootstrapping assuming multinominal distribution (Sison and Glatz, 1995). R (R Core Team, 2021) with the package DescTools (Signorell et mult. al., 2021) was used for calculation.

	total		non SDE		SDE	
	lower	upper	lower	upper	lower	upper
aluminosilicates / Ałrich	0.432	0.567	0.295	0.464	0.643	0.83
carbonates	0.03	0.165	0.023	0.192	0	0.187
silicon dioxide	0	0.105	0	0.123	0	0.144
biological	0	0.05	0	0.107	0	0.101
soot	0	0.08	0	0.092	0	0.13
C-rich	0.05	0.185	0.085	0.255	0	0.13
metal oxides	0	0.115	0	0.154	0	0.116
sulfates	0	0.09	0	0.115	0	0.116
mixtures	0.06	0.195	0.101	0.27	0	0.13
no classification possible	0	0.115	0	0.146	0	0.13

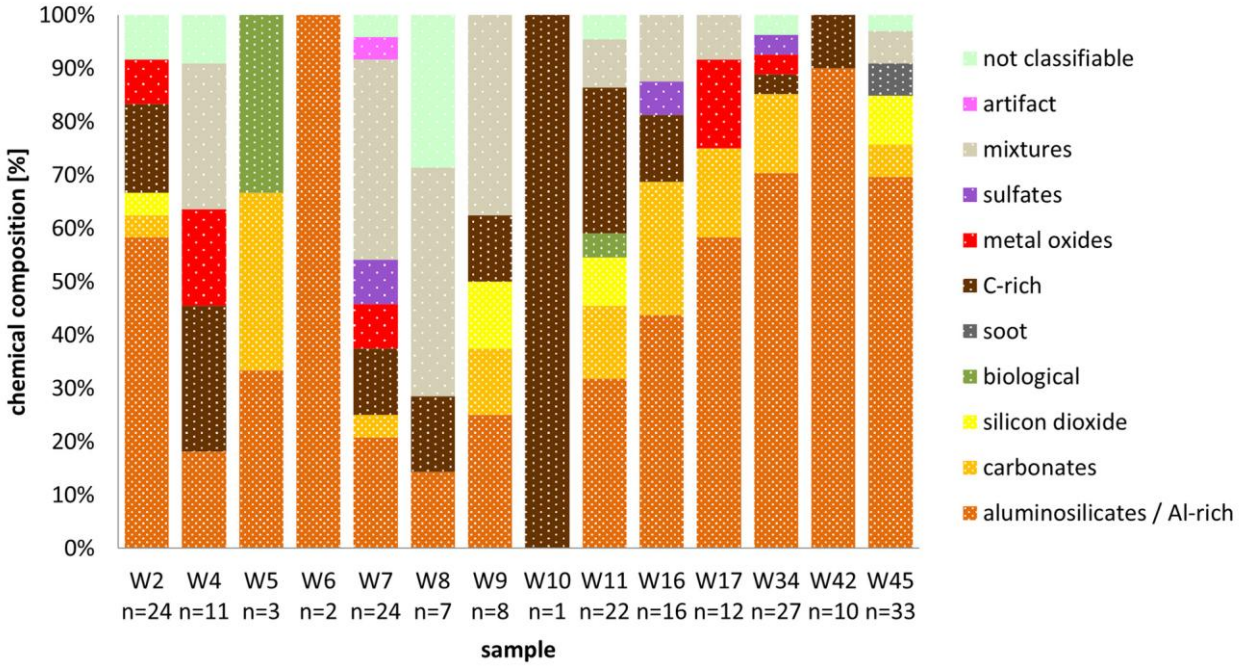


Figure S4: Chemical composition of individual samples from CLACE/INUIT 2017 at the high-altitude research station Jungfraujoch (activated at T = -30°C and RH = 99 / 101 %; RH = 95 / 97% was chosen for W16 due to cluster formation at higher RH) with the number of particles identified per sample (n).

Table S2: 0.95 confidence intervals for the chemical composition shown in Fig. 12, calculated by bootstrapping assuming multinominal distribution (Sison and Glatz, 1995). R (R Core Team, 2021) with the package DescTools (Signorell et mult. al., 2021) was used for calculation.

	0.3 - 0.5 µm		0.5 – 1 µm		1 – 2 µm		2 – 3 µm		3 – 4 µm	
	lower	upper	lower	upper	lower	upper	lower	upper	lower	upper
aluminosilicates / Al-rich	0	0.513	0.1773	0.86	0.342	0.783	0.361	0.9	0.326	0.874
carbonates	0	0	0	0.166	0	0.272	0	0.278	0	0.15
silicon dioxide	0	0.9514	0	0	0	0.131	0	0.178	0	0
biological	0	0	0	0	0	0	0	0	0	0
soot	0	0	0	0.166	0	0	0	0	0	0
C-rich	0	0.513	0	0.3907	0	0.272	0	0	0	0.232
metal oxides	0	0.763	0	0.3257	0	0.131	0	0	0	0
sulfates	0	0	0	0	0	0	0	0.278	0	0.15
mixtures	0	0	0	0.166	0	0.206	0	0.278	0.001	0.479
no classification possible	0	0	0	0.3257	0	0.084	0	0	0	0

	4 – 5 µm		5 – 6 µm		6 - 34.3 µm		size not meas.	
	lower	upper	lower	upper	lower	upper	lower	upper
aluminosilicates / Al-rich	0.078	0.745	0	0.0769	0.273	0.727	0	0
carbonates	0	0.336	0	0.507	0	0.285	0	0
silicon dioxide	0	0.218	0	0	0	0	0	0
biological	0	0	0	0	0	0.154	0	0
soot	0	0	0	0	0	0	0	0.513
C-rich	0	0.336	0	0.333	0	0.323	0	0.513
metal oxides	0	0.336	0	0	0	0	0	0
sulfates	0	0	0	0.333	0	0	0	0
mixtures	0	0.435	0	0.648	0	0.323	0	0
no classification possible	0	0	0	0	0	0	0.38	1

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Sison, C. P. and Glaz, J.: Simultaneous Confidence Intervals and Sample Size Determination for Multinomial Proportions, Journal of the American Statistical Association, 90, 366-369, doi: 10.1080/01621459.1995.10476521, 1995