Figure S1. Temperature difference between the different wells of PCR1 and PCR2 and the aluminum block.
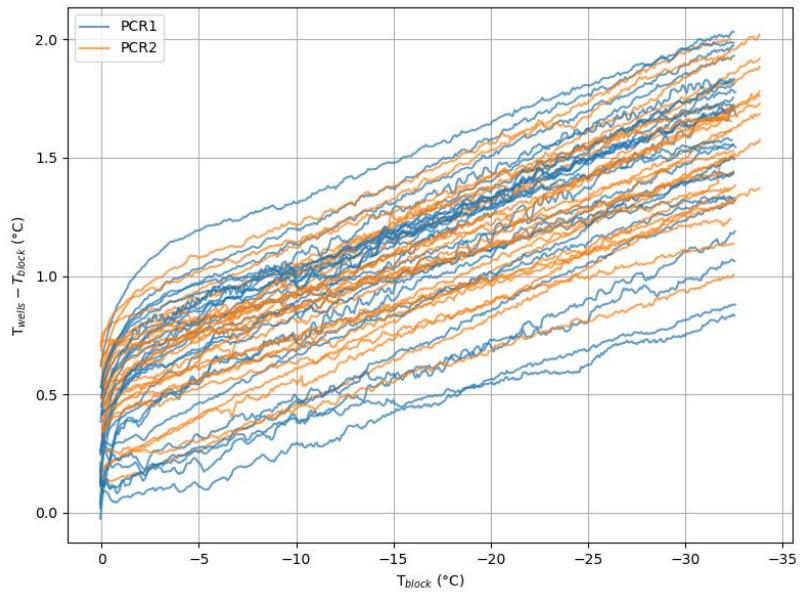
Figure S2. Temperature difference between PCR 1 (blue) and PCR 2 (orange) and the aluminum block.
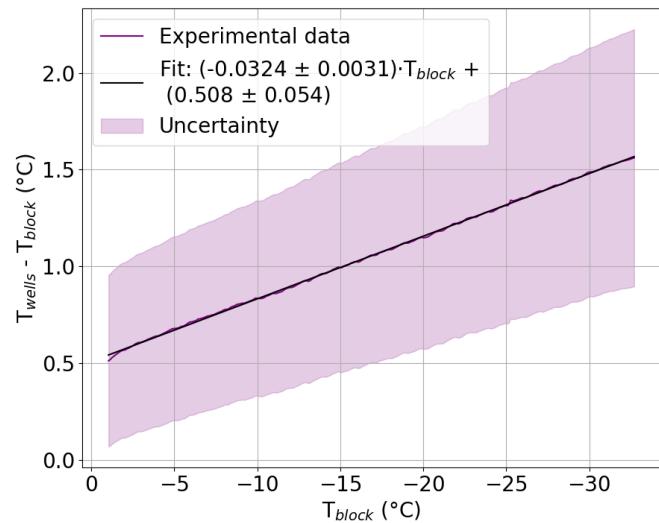
Figure S3. Temperature calibration of GRAINS.
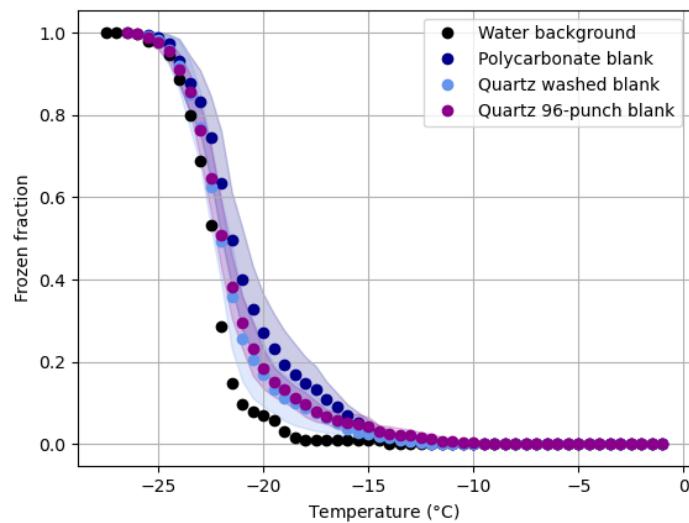
Figure S4. Frozen fraction of the different filter backgrounds for each method. Data shown correspond to averages from 6 different experiments, where shaded area corresponds to the standard deviation.

Section	Purpose	Method / Setup	Key Details
Validation with NX Illite	Test reproducibility with reference dust	*Wet suspension*: mix NX Illite powder ($1-10^{-3}$ g L^{-1}) directly in water. *Dry dispersion*: aerosolize NX Illite (SwisensAtomizer), sample on 25 mm polycarbonate filters at 5 Lmin^{-1} for 90 minutes. Extraction of particles by manual agitation for 60 s.	APS (0.5–20 µm) used to determine size distribution & surface area; density = 2.65 g cm^{-3}, shape factor = 1.49.
Intercomparison with FrESH & INSEKT	Compare GRAINS with other INP devices	Ambient samples: GRAINS vs FrESH: Finland samples, 24 h sampling at 16.6 Lmin^{-1}. Extraction of particles with a vortex agitator for 30 seconds for FrESH, manual agitation during 60 seconds for GRAINS. GRAINS vs INSEKT: 24 h sampling at 5 Lmin^{-1}. Polycarbonate filters pre-cleaned (10% H_2O_2). Extraction of particles with agitating rotator during 20 min. AIDAd samples: K-feldspar, Arizona Test Dust (ATD), Soil Dust South Africa (SDSA01) and deposited Saharan Dust (SD). Injected into AIDAd using a 2.5 µm cutoff cyclone, 60 minutes sampling at 5 Lmin^{-1}. Polycarbonate filters pre-cleaned (10% H_2O_2). Extraction of particles with agitating rotator during 20 min.	Finland samples: half filters split & analyzed in both labs, 10-fold dilutions. KIT samples: frozen transport/storage, 15- and 225-fold dilutions. AIDAd samples: frozen transport/storage, 10- and 100-fold dilutions. Simultaneous PINE measurements.
Testing substrates & methods	Evaluate different filter substrates & particle extraction protocols	Sampling at UGR (urban background) for 24 h every 4 days: 47 mm polycarbonate filters at 9 L min^{-1} (no-size cut). 150 mm quartz filters at 500 L min^{-1} (PM$_{10}$ inlet).	Polycarbonate filters: stored frozen, analyzed by washing in 20 mL water, manual agitation for 60 s. Quartz filters: pre-heated (205 °C), punched & analyzed two ways: **96-punch** (1 mm punches into wells, 100 µL water each) and **Punch-washed** (1 cm punch washed in 20 mL water, manual agitation for 60 s).

Table S1. Summary of the different methodology subsections.
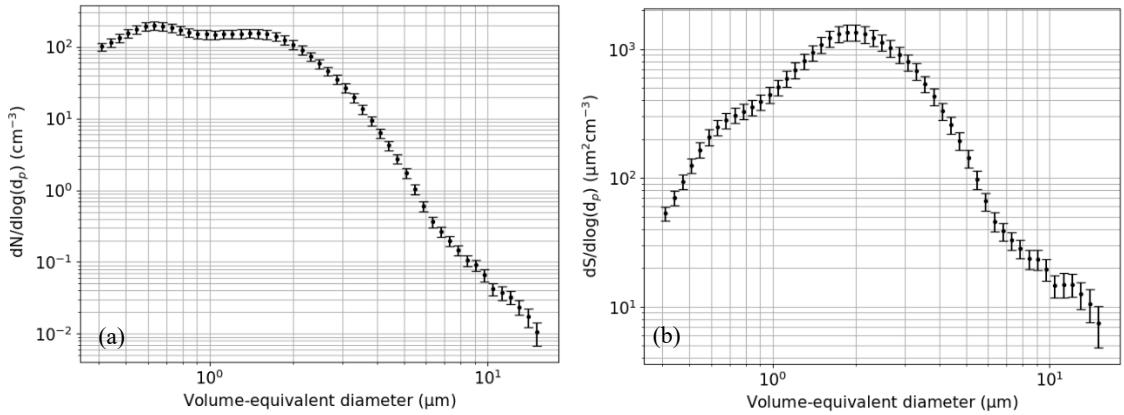
Figure S5. Average number size distribution (a) and surface size distribution (b) measured with the APS for NX Illite. Conversion from aerodynamic diameter to volume-equivalent diameter assuming a density of 2.65 gcm[-3] and a dynamic shape factor of 1.49. Errorbars shown correspond to the standard error of the average.
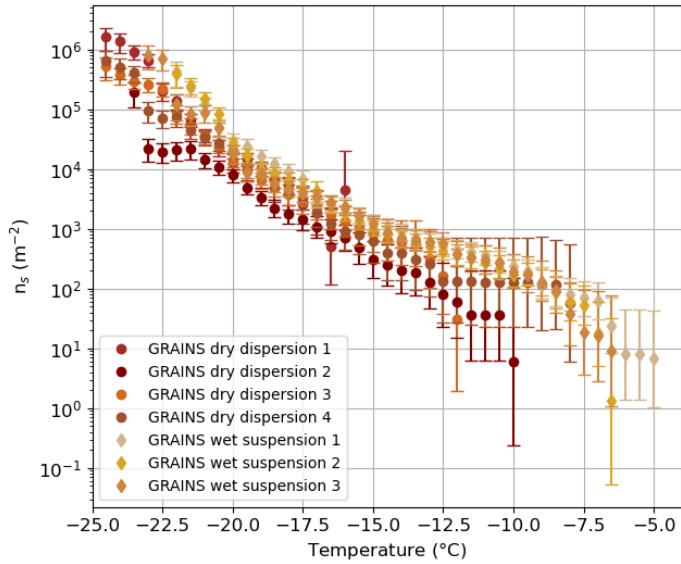
Figure S6. Ns(T) of NX Illite obtained with the GRAINS instrument. Uncertainties were calculated based on Agresti and Coull (1998).
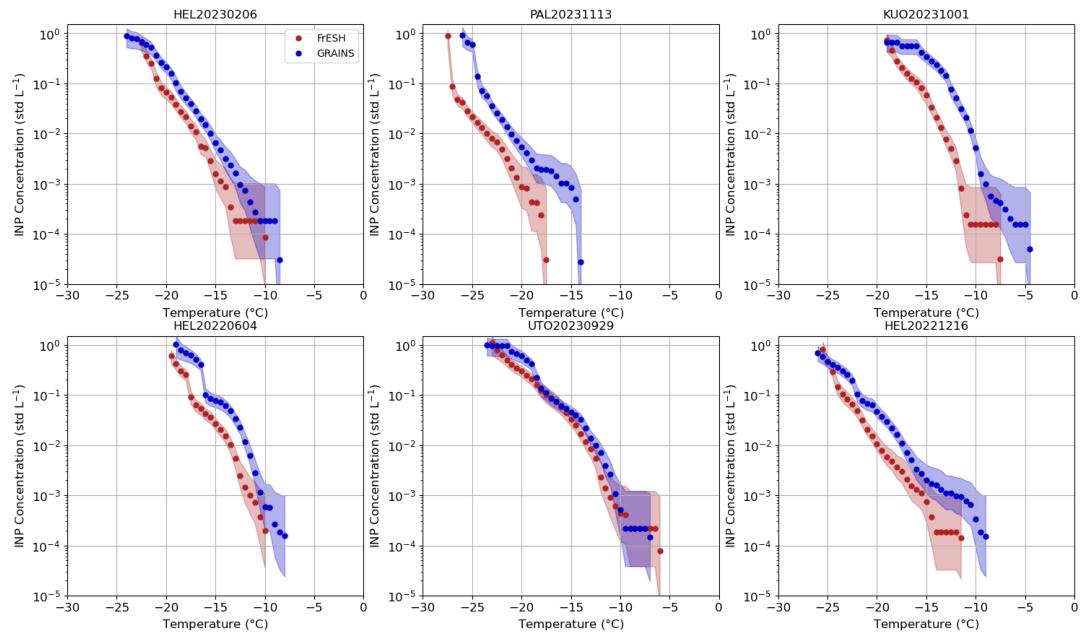
Figure S7. INP concentration of the FMI filters analyzed by GRAINS (blue) and FrESH (red). Shaded area represents uncertainty calculated based on Agresti and Coull (1998).
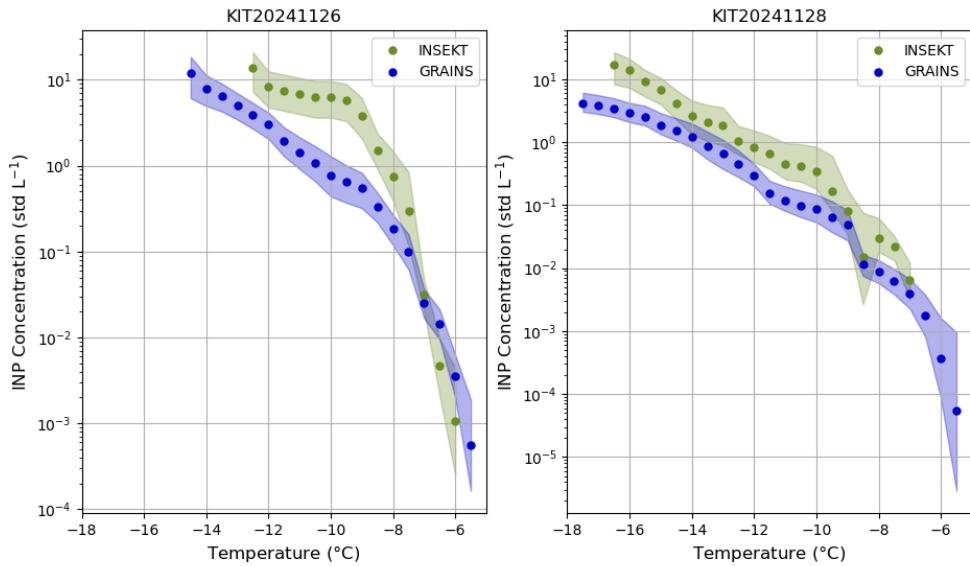
Figure S8. INP concentration of the KIT filters analyzed by GRAINS (blue) and INSEKT (green). Shaded area represents uncertainty calculated based on Agresti and Coull (1998).
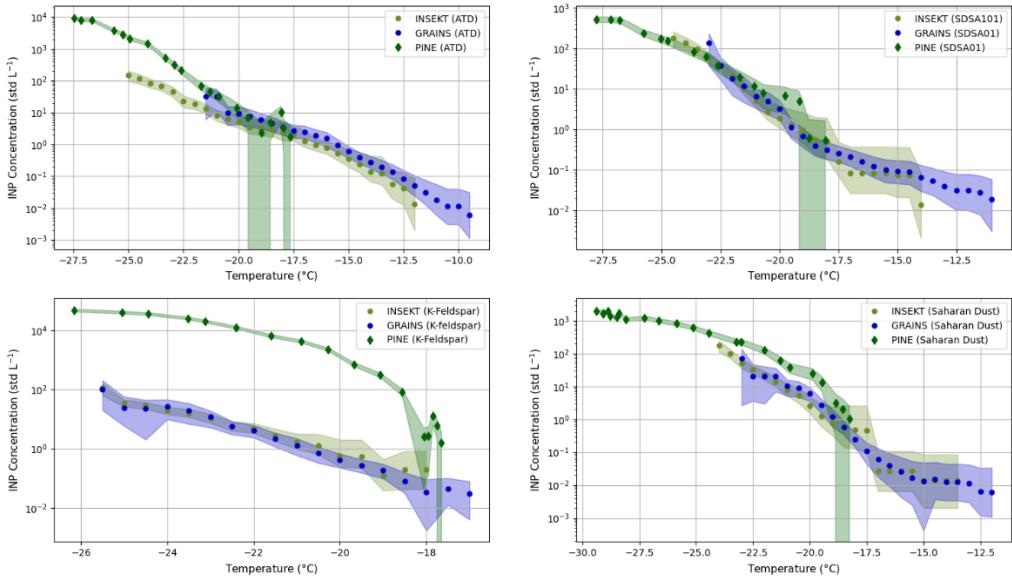
Figure S9. INP concentration of the experiments conducted at the AIDAd chamber; sampled filters are analyzed by GRAINS and INSEKT and PINE measurements were conducted in-situ at AIDAd. Shaded area for INSEKT and GRAINS represents uncertainty calculated based on Agresti and Coull (1998), whereas for PINE it represents a combination of a 10% of the INP concentration and the square root of the number of ice crystals.
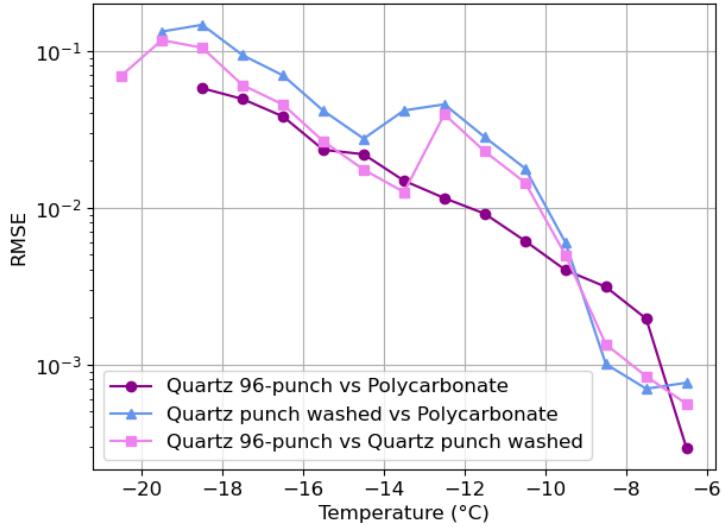
Figure S10. Relationship of the root mean squared error (RMSE) between the three methods with temperature.
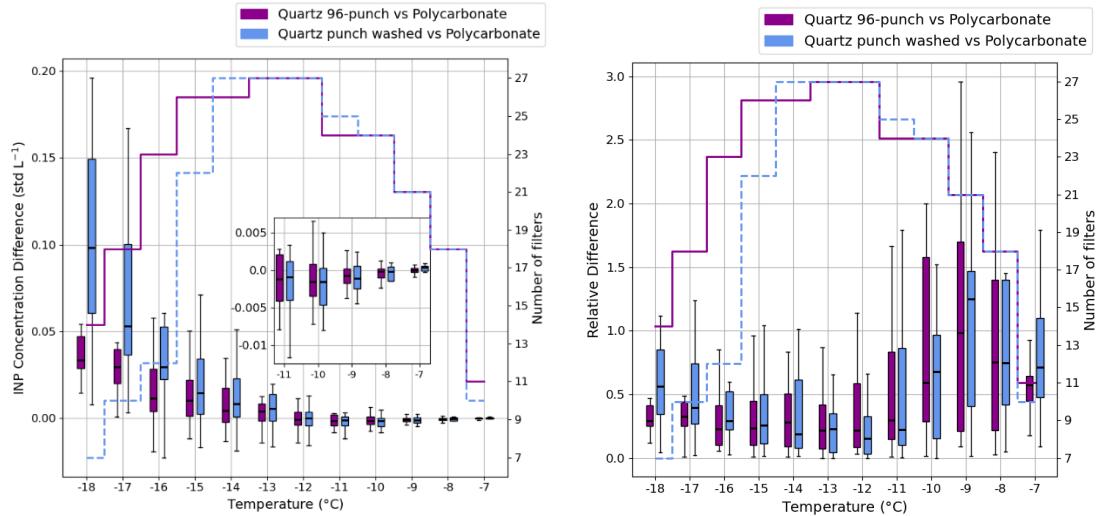
Figure S11. Boxplots of the INP concentration absolute (left) and relative (right) differences between methods at twelve different temperatures (from -18 ºC to -7 ºC). Dashed lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) quartiles, where the difference between them represents the inter-quartile range (IQR). Lower and upper whiskers show Q1-1.5IQR and Q3+1.5IQR, respectively. Outliers are not shown for clarity. Right axis represents the number of filters (solid and dashed lines) used for the statistics shown in the Figure, whereas the inset plot in the left subfigure corresponds to a zoom of the boxplots between -7ºC and -11ºC for better visualization.