



Supplement of

Evaluation of different sampling methods to determine the ice-nucleating particle concentration in the atmosphere using the GRANada Ice Nuclei Spectrometer (GRAINS)

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Figure S1. Set up of the GRAINS instrument.

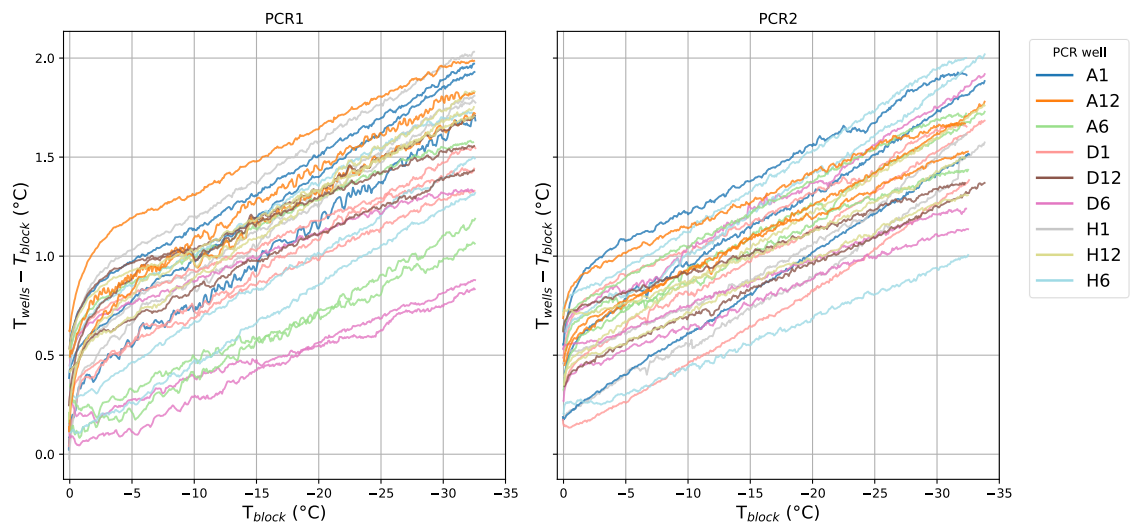


Figure S2. Temperature difference between the different wells of PCR1 and PCR2 and the aluminum block.

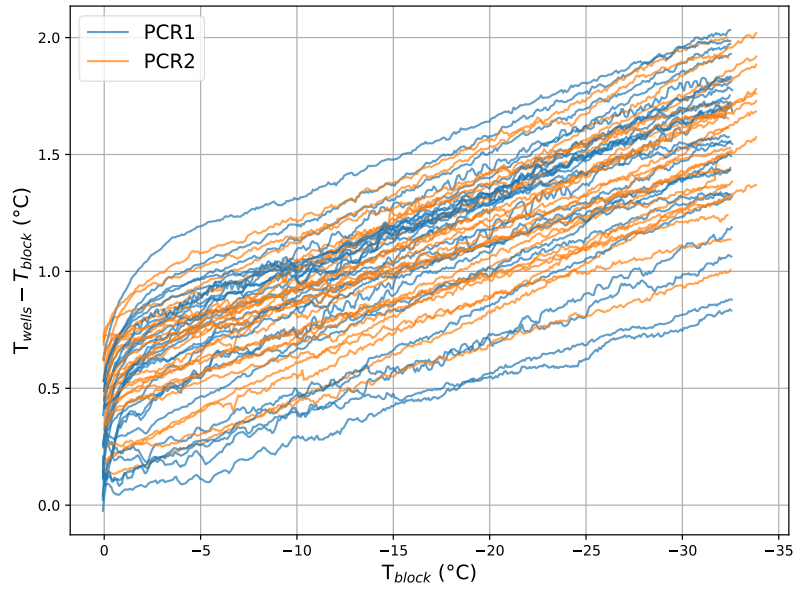


Figure S3. Temperature difference between PCR 1 (blue) and PCR 2 (orange) and the aluminum block.

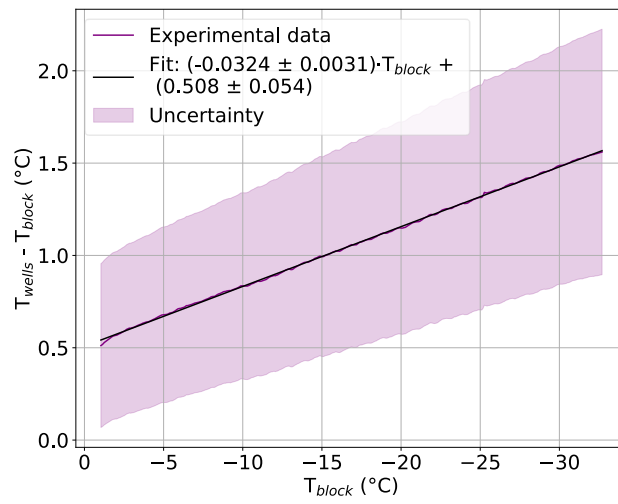


Figure S4. Temperature calibration of GRAINS.

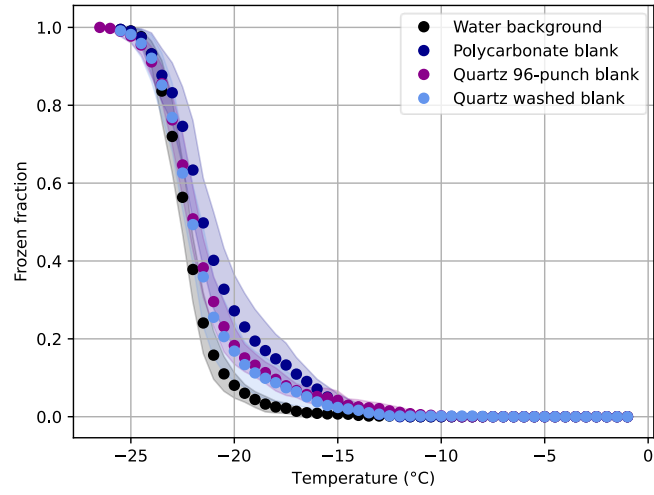


Figure S5. Frozen fraction of the different filter backgrounds for each method. Data shown correspond to averages from 6 different experiments, where shaded area corresponds to the standard deviation.

Section	Purpose	Method / Setup	Key Details
Validation with NX Illite	Test reproducibility with reference dust	<p><i>Wet suspension:</i> mix NX Illite powder ($1-10^{-3}$ g L⁻¹) directly in water.</p> <p><i>Dry dispersion:</i> 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.</p>	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	<p>Ambient samples:</p> <p>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.</p> <p>GRAINS vs INSEKT: 24 h sampling at 5 Lmin⁻¹. Polycarbonate filters pre-cleaned (10% H₂O₂). Extraction of particles with agitating rotator during 20 min.</p> <p>AIDAd samples:</p> <p>K-feldspar, Arizona Test Dust (ATD), Soil Dust South Africa (SDSA01) and deposited Saharan Dust (SD).</p> <p>Injected into AIDAd using a 2.5 μm cutoff cyclone, 60 minutes sampling at 5 Lmin⁻¹. Polycarbonate filters pre-cleaned (10% H₂O₂). Extraction of particles with agitating rotator during 20 min.</p>	<p>Finland samples: half filters split & analyzed in both labs, 10-fold dilutions.</p> <p>KIT samples: frozen transport/storage, 15- and 225-fold dilutions.</p> <p>AIDAd samples: frozen transport/storage, 10- and 100-fold dilutions. Simultaneous PINE measurements.</p>
Testing substrates & methods	Evaluate different filter substrates & particle extraction protocols	<p>Sampling at UGR (urban background) for 24 h every 4 days:</p> <p>47 mm polycarbonate filters at 9 L min⁻¹ (no-size cut).</p> <p>150 mm quartz filters at 500 L min⁻¹ (PM₁₀ inlet).</p>	<p>Polycarbonate filters: stored frozen, analyzed by washing in 20 mL water, manual agitation for 60 s.</p> <p>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).</p>

Table S1. Summary of the different methodology subsections.

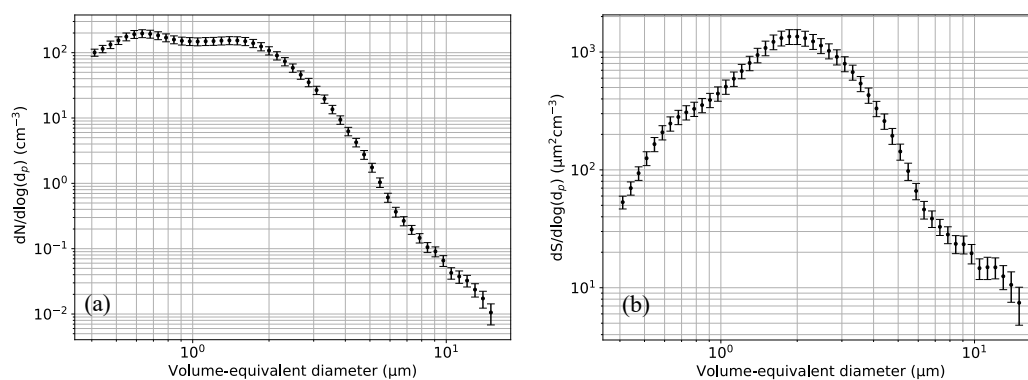


Figure S6. Average number size distribution (a) and surface size distribution (b) measured with the APS for NX Illite. Conversion from aerodynamic diameter to volume-equivalent diameter assuming a density of 2.65 g cm^{-3} and a dynamic shape factor of 1.49. Errorbars shown correspond to the standard error of the average.

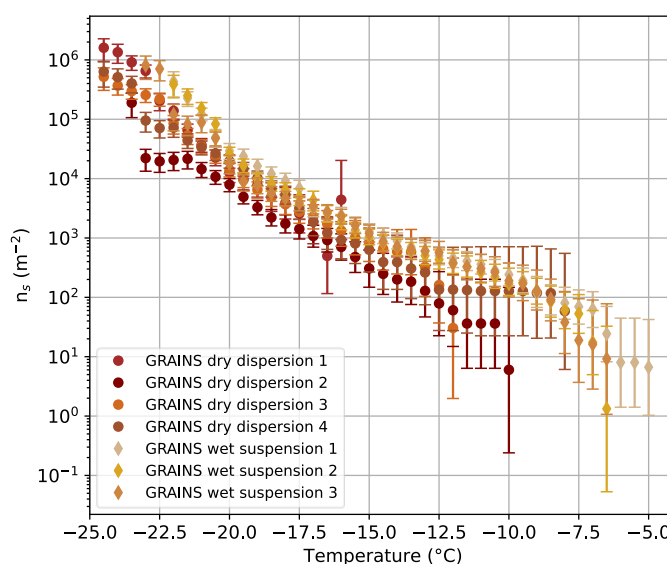


Figure S7. $n_s(T)$ of NX Illite obtained with the GRAINS instrument. Uncertainties were obtained by evaluating the analytical expression of the n_s at the lower and upper confidence limits of the frozen fraction, which were calculated based on Agresti and Coull (1998).

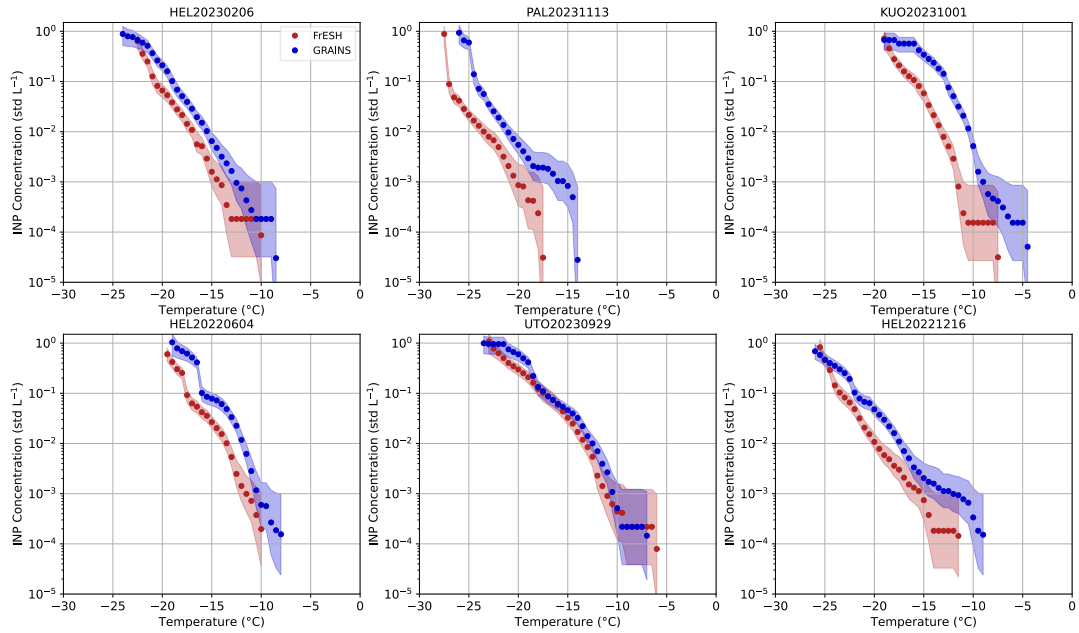


Figure S8. INP concentration of the FMI filters analyzed by GRAINS (blue) and FrESH (red). Shaded area represents uncertainty calculated based on Agresti and Coull (1998).

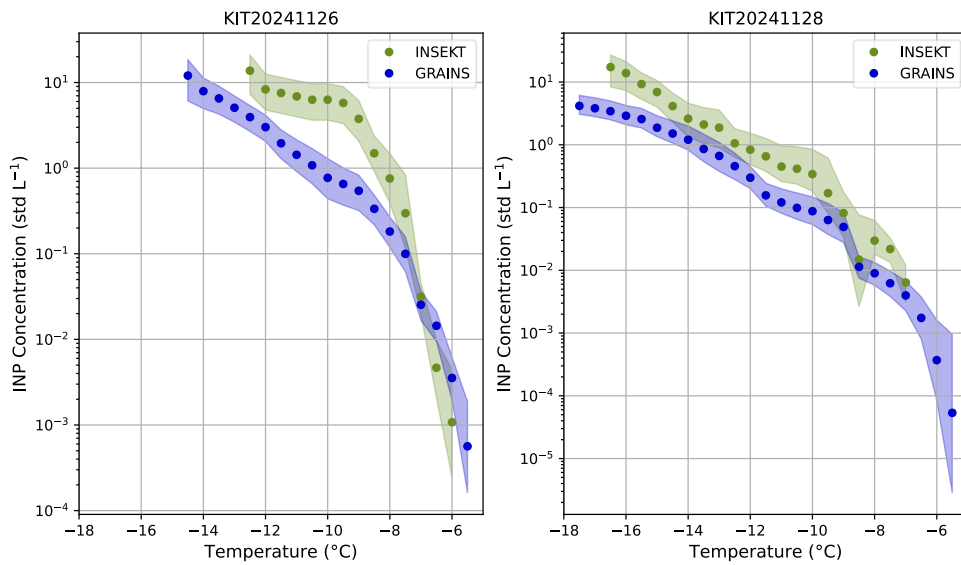


Figure S9. INP concentration of the KIT filters analyzed by GRAINS (blue) and INSEKT (green). Shaded area represents uncertainty calculated based on Agresti and Coull (1998).

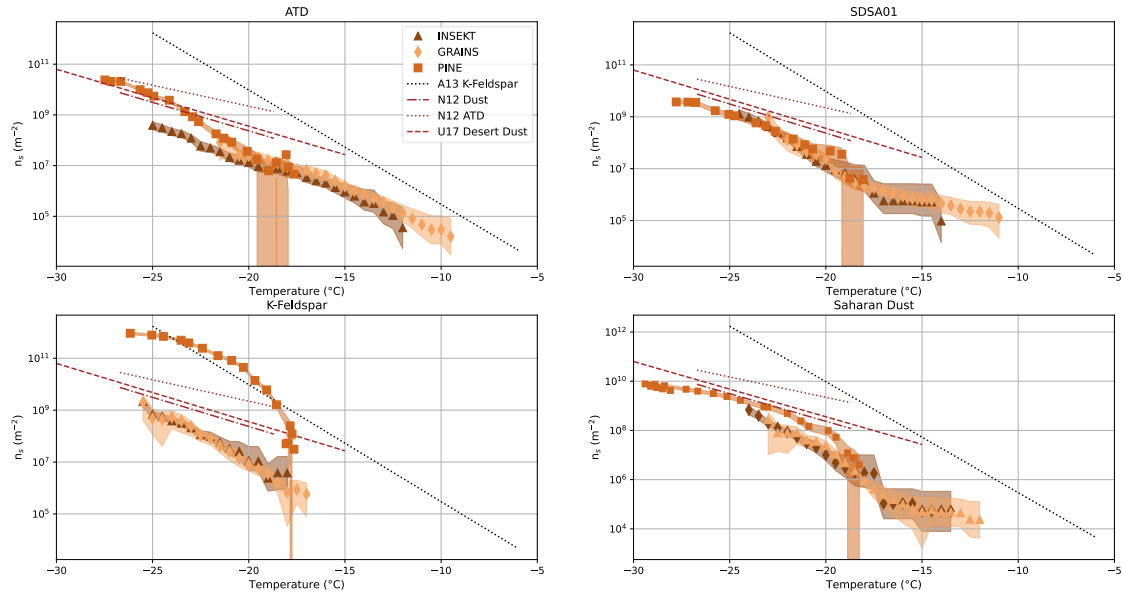


Figure S10. Ice nucleation active sites (INAS) density (n_s) per unit of aerosol surface obtained with INSEKT, GRAINS and PINE during the experiments conducted at AIDA. Surface area was obtained as the lognormal fit of the size distributions obtained with the combination of the Scanning Mobility Particle Sizer (SMPS) and Aerodynamic Particle Sizer (APS). Shaded area represents uncertainty following error propagation, where the error of the frozen fractions for INSEKT and GRAINS was obtained by evaluating the analytical expression of the n_s at the lower and upper confidence limits of the frozen fraction, which were on Agresti and Coull (1998), whereas for PINE it was calculated as a combination of a 10% of the INP concentration and the square root of the number of ice crystals. Parametrizations from Atkinson et al. (2013) (A13), Niemand et al. (2012) (N12) and Ulrich et al. (2017) (U17) are also shown.

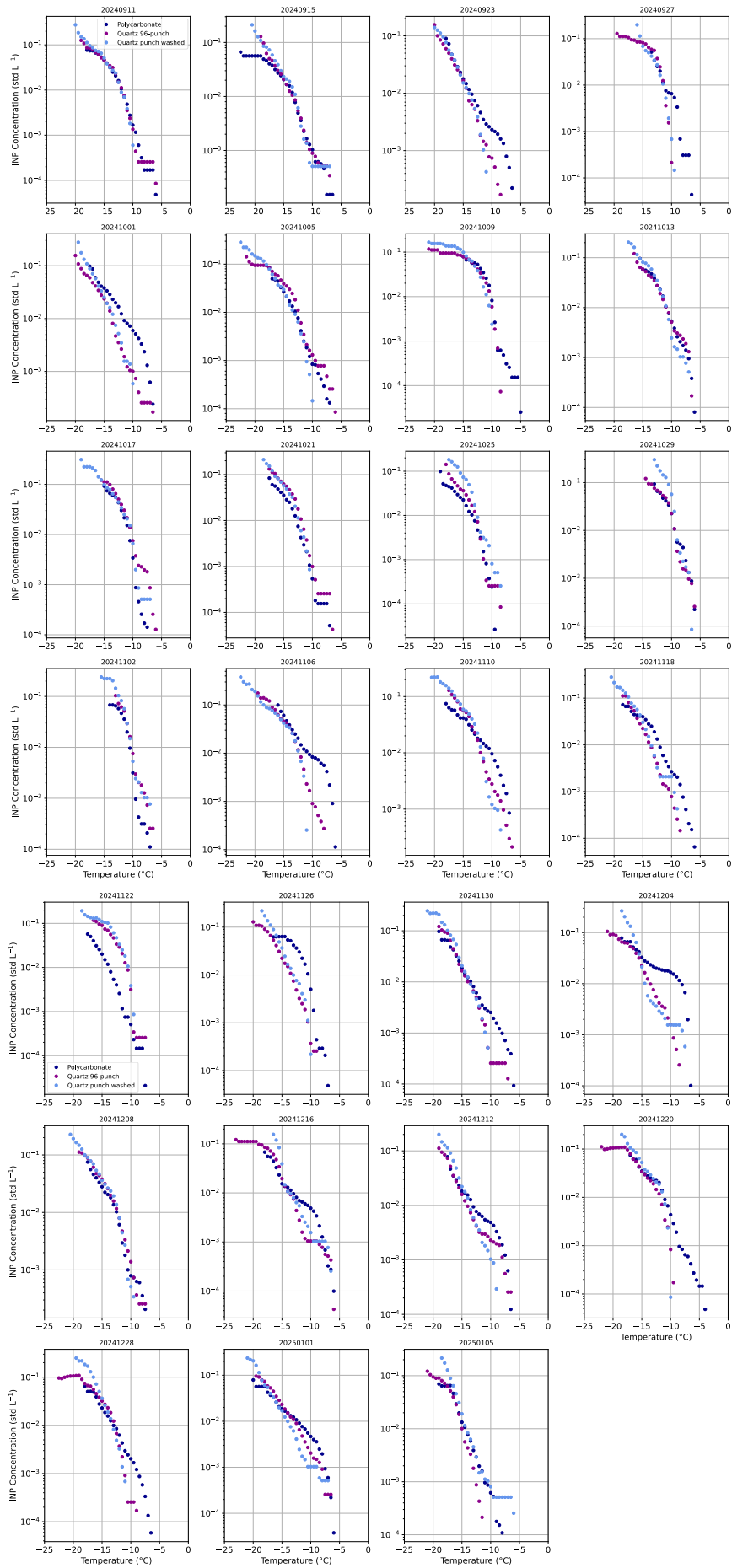


Figure S11. Individual spectra for the comparison of the different methods for sampling and analyzing INP. Uncertainties are not shown for clarity.

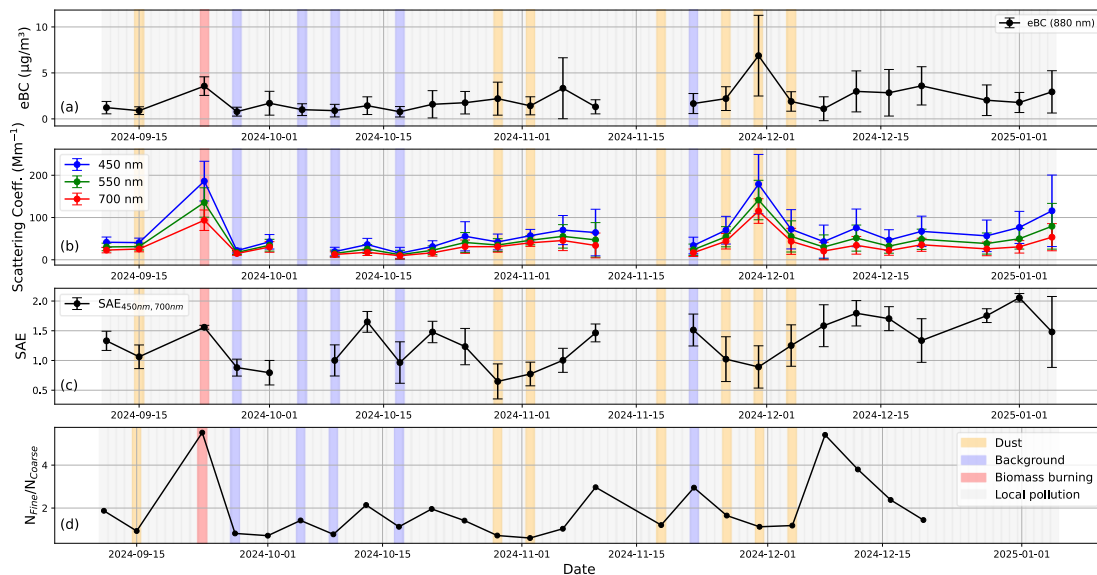


Figure S12. (a) Time series of the daily averages of the equivalent black carbon mass concentration (eBC) measured at 880 nm with an aethalometer (AE33, Aerosol Magee Scientific). (b) Integrated aerosol light-scattering coefficients at 450, 550 and 700 nm measured with a nephelometer (3563, TSI Inc.). (c) Scattering Angstrom Exponent (SAE) between 450 nm and 700 nm. (d) Ratio of particle number concentration of fine (diameter < 1 μm) and coarse (diameter > 1 μm) particles ($N_{\text{Fine}}/N_{\text{Coarse}}$) measured with an Aerodynamic Particle Sizer (APS, 3321, TSI Inc.). Error bars represent the standard deviation of the daily averages and shaded areas represent different aerosol conditions during each day.

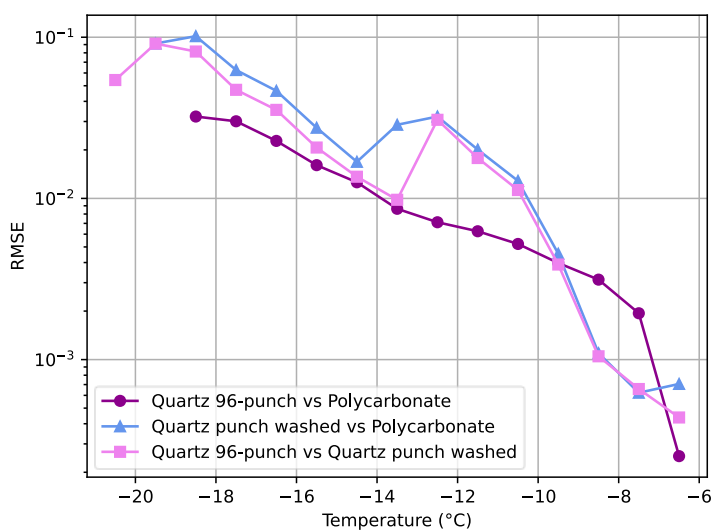


Figure S13. Relationship of the root mean squared error (RMSE) between the three methods with temperature.

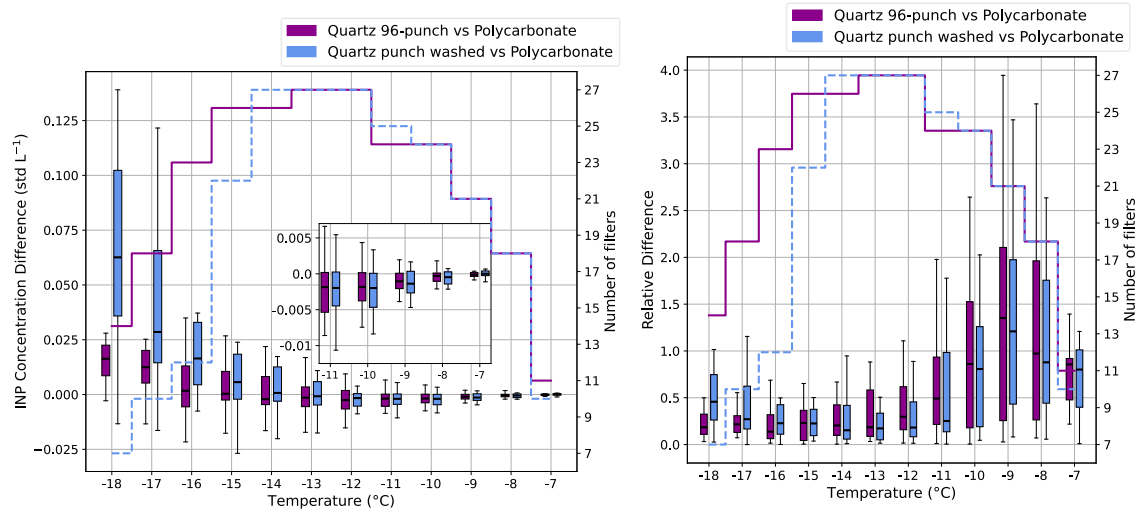


Figure S14. Boxplots of the INP concentration absolute (left) and relative (right) differences between methods at twelve different temperatures (from -18 °C to -7 °C). Dashed lines represent the median values, lower and upper limits in the boxplots correspond to the first (Q1) and third (Q3) quartiles, 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 -7°C and -11°C for better visualization.