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Supplement of

Measurement report: High Arctic aerosol hygroscopicity at sub- and supersaturated conditions during spring and summer

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Supplement

Supplementary material 1

Calibration of the CCN counter.

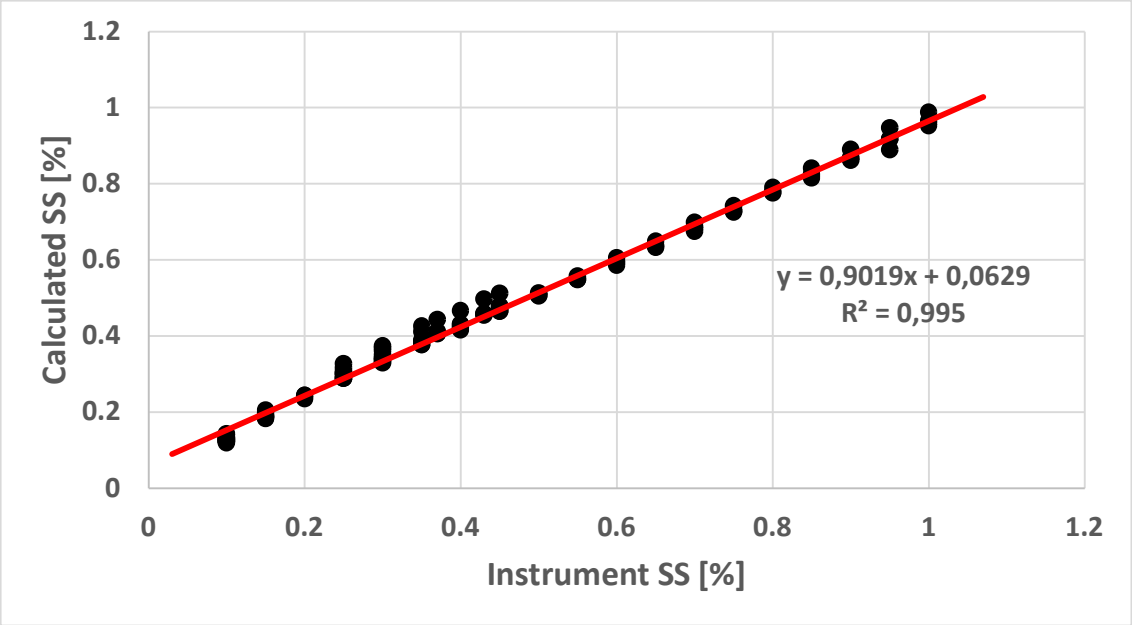

Figure S1. CCN counter calibration curve. Calculated supersaturation as determined by the E-AIM model and Köhler equation, as a function of SS set on the instrument. The calibration is a composite of all four performed CCNC calibrations before, during, and after the field studies.

Supplementary material 2

Calculations on non-isokinetic sampling.

The relation of the flow velocity in the sample duct (U_0) to the sample flow velocity (U) is given by:

$$\frac{U_0}{U} = \frac{\frac{V_0}{2\pi \cdot r_0^2}}{\frac{V}{2\pi \cdot r^2}} \quad \text{(S1)}$$

Where V_0 is the volumetric flow in the sample duct, and r_0 is the tube radius of the sample duct, V is the sample volumetric flow and r the sample tube radius. V_0 = 4 m^3 min^{-1}, r_0 = 0.05 m, V = 0.0015 m^3 min^{-1} and r = 0.002 m, yielding $\frac{U_0}{U} = 4.3$.

The Stokes number (*stk*) of an aerosol particle is:

$$stk = \frac{Dp^2 \cdot \rho \cdot C_C \cdot U_0}{18\eta \cdot D_{char}} \quad \text{(S2)}$$

Where D_p is the particle diameter, ρ the particle density, C_c the Cunningham slip correction factor, η is the gas viscosity and D_{char} the characteristic dimension of the system (sample tube diameter).

The upper end of the SMPS measurement range is 900 nm, thus we use D_p = 1000 nm, ρ = 1300 kg m^{-3}, C_c(1000 nm) = 1.15, η = 0.0000181 kg m^{-1} s^{-1} and D_{char} is the sample tube diameter (2* r_0), yielding $stk_{1000} = 0.09$. Assuming D_p = 250 nm and the corresponding C_c(100 nm) = 2.93, yielding $stk_{250} = 5.5 \cdot 10^{-3}$. Taking the square root of these Stokes number yields $\sqrt{stk_{1000}} = 0.30$ and $\sqrt{stk_{250}} = 0.07$, respectively.

Hinds (1999) displays Figure 10.4 on page 211 where the effect of non-isokinetic sampling is illustrated, as a function of $\sqrt{stk}$. By reading the figure, it is seen that although we did sample non-isokinetically, oversampling of particles is negligible for sub-micrometer particles.

Supplementary material 3

Correction of CCN measurements

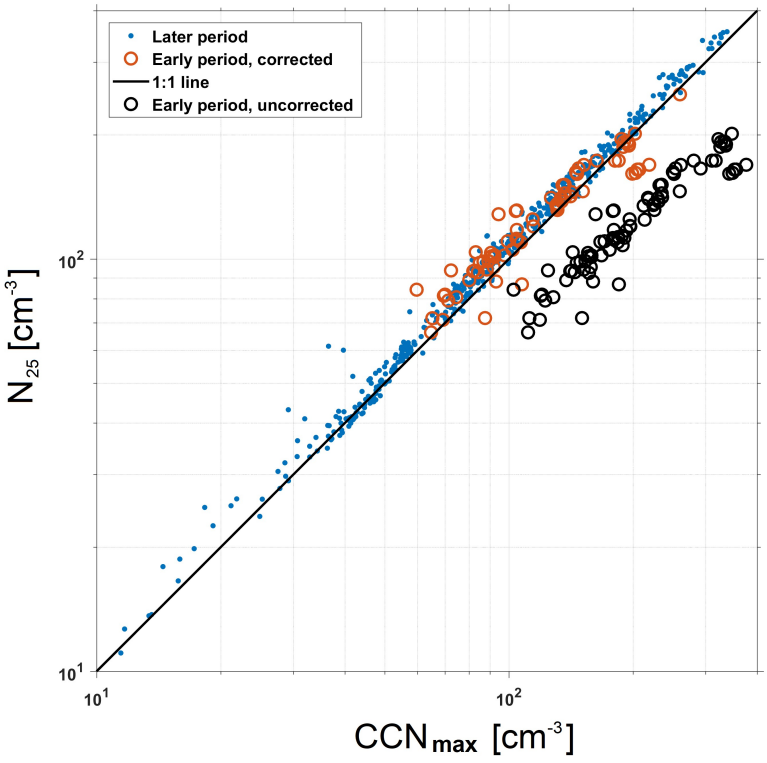

Figure S2. Correction of CCN measurements during 20-30 April of 2016. N_{25} particle concentration is shown as a function of CCN_{max}. Blue dots show measurements later than 30 Apr, black circles uncorrected measurements before 30 Apr, and brown circles are the corrected measurements after 30 Apr. The correction factor of 0.582 was applied to all CCN measurements recorded before 30 Apr.

Supplementary material 4

HTDMA calibration measurements

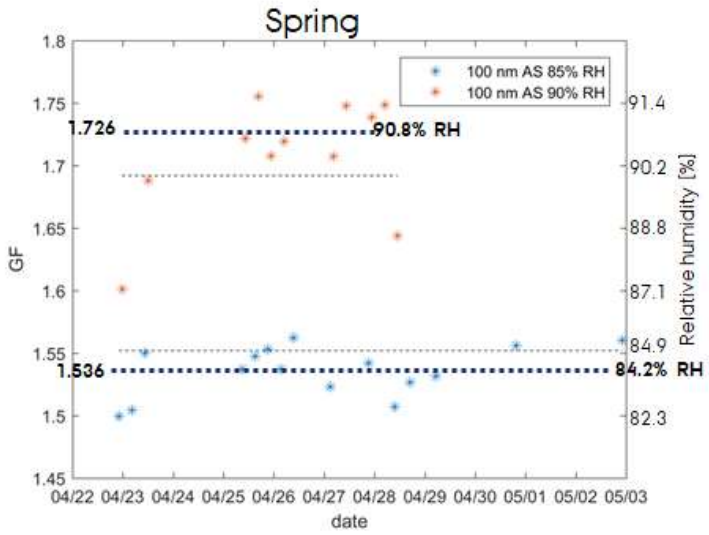

Figure S3. Measurements of GF of 100 nm monodisperse ammonium sulfate aerosol during the spring measurement period. Blue stars are measurements at 85% RH, red stars at 90% RH. Thick dotted lines are the resulting average GFs, thin dotted lines show the expected correct GF as calculated from the E-AIM model and Köhler equation. Y-axes show both GF and corresponding RH, such that the RH actually present inside the HTDMA instrument can be directly read.

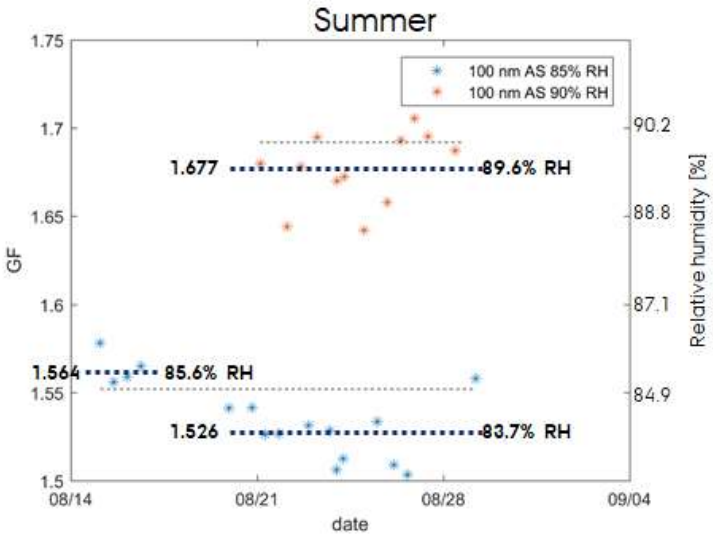

Figure S4. Measurements of GF of 100 nm monodisperse ammonium sulfate aerosol during the summer measurement period. Blue stars are measurements at 85% RH, red stars at 90% RH. Thick dotted lines are the resulting average GFs, thin dotted lines show the expected correct GF as calculated from the E-AIM model and Köhler equation. Y-axes show both GF and corresponding RH, such that the RH actually present inside the HTDMA instrument can be directly read. Note that two different average GF values are used for 85% RH.

Supplementary material 5

Correction of HTDMA offsets of RH and re-calculation of GF at 85 and 90% RH, respectively.

The κ-Köhler equation for particle GF measurements is:

$$\frac{RH}{\exp\left(\frac{A}{Dp_{dry} \cdot GF}\right)} = \frac{GF^3 - 1}{GF^3 - (1 - \kappa_{HTDMA})} \quad \text{(S3)}$$

The equation is slightly rearranged:

$$\frac{RH}{GF^3 - 1} GF^3 - (1 - \kappa_{HTDMA}) = \exp\left(\frac{A}{Dp_{dry} \cdot GF}\right) \quad \text{(S4)}$$

The left-hand side is defined as an arbitrary variable, x, such that the following holds:

$$GF = \frac{A}{Dp_{dry} \cdot \ln(x)} \quad \text{(S5)}$$

The initial GF measured at the slightly offset RH_0 is defined as GF_0, and GF_i is determined from the calculations. RH_i is the relative humidity at which GF_i is determined. Now the following calculations can be run iteratively with RH_i as either 85 or 90% RH:

$$\frac{RH}{GF_0^3 - 1} GF_0^3 - (1 - \kappa_{HTDMA}) = x \quad \text{(S6)}$$

$$GF_i = \frac{A}{Dp_{dry} \cdot \ln(x)} \quad \text{(S7)}$$

$$GF_0 = GF_i \quad \text{(S8)}$$

GF_0 and GF_i converge quickly and usually 100 iterations are more than sufficient.

We used Matlab® to perform these calculations, any other programming- or scripting language can be used, even Excel®.

Supplementary material 6

Estimation of uncertainty on CCN and GF measurements.

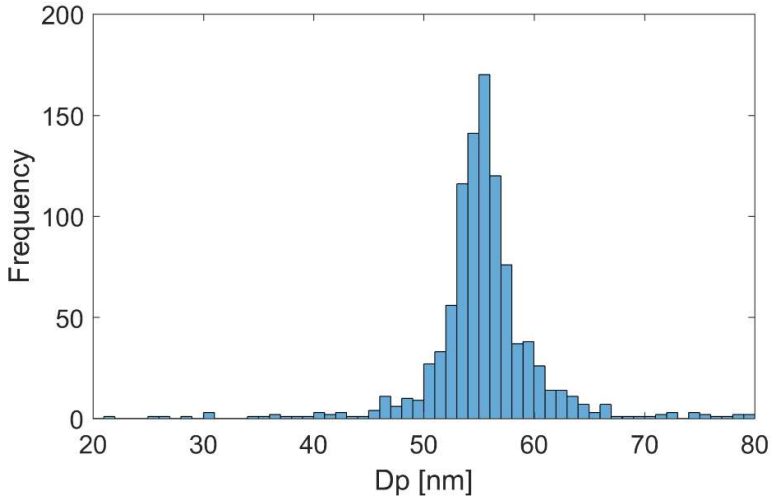

Figure S5. Example histogram resulting from iteratively based error estimation. In this case, Dp_{crit} is estimated based on random errors of CCN measurements and size distributions. An automatically fit normal distribution gives the final estimate and standard deviation.

Supplementary material 7

Ambient aerosol number size distribution.

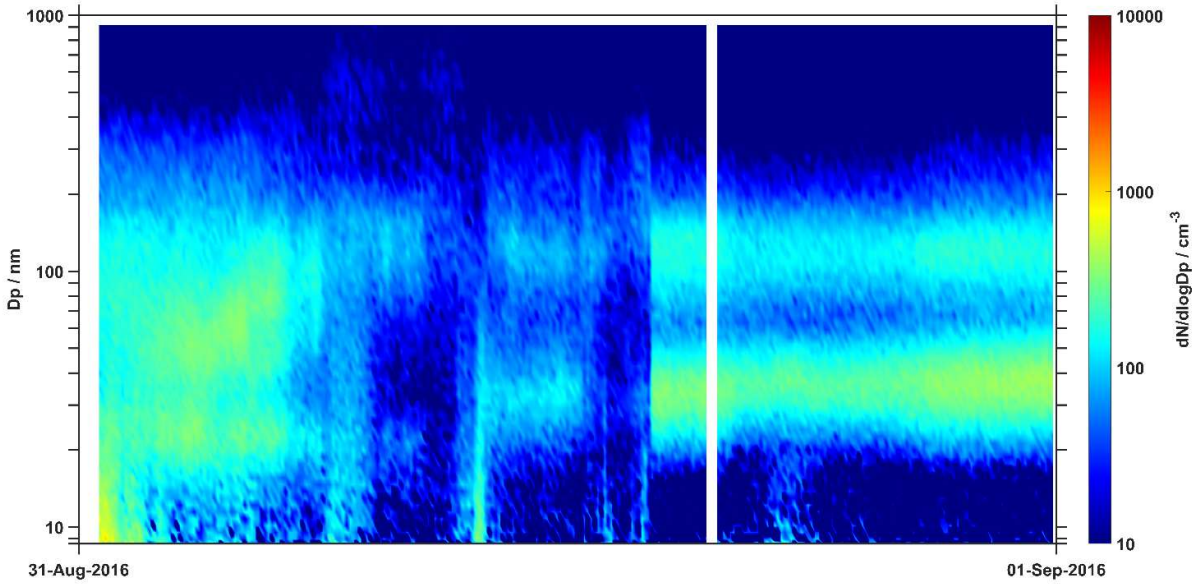

Figure S6. Ambient aerosol number size distribution at Villum from 31 Aug 2016.

Reference

Hinds, W. C.: Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles, Wiley, 1999.