

Derivation of the ice nucleating particle (INP) concentration

Immersion mode INP measurements are performed by sampling at 243 K and saturation ratio with respect to water (S_w) of 1.04. At these conditions, ambient Cloud Condensation Nuclei (CCN) should activate to supercooled droplets, and the fraction of which are INPs form ice crystals. The size of the hydrometeors exiting the chamber is used to differentiate between liquid droplets and ice crystals because ice grows to larger sizes at the prevailing conditions. The survival rate of ambient particles $\geq 2 \mu\text{m}$ through the tubing and the warm, dry chamber on the JFJ is 33%. No ambient particles $\geq 3 \mu\text{m}$ survived. Differential measurements at the total aerosol inlet with a hand-held Optical Particle Counter (OPC) and after the chamber were used to calculate the particle loss rates.

Diffusional growth calculations at the described conditions, taking into account the supersaturation equilibration within the chamber predicts hydrometeors to grow to a maximum size of $D = 3.31 \mu\text{m}$ (Zurich, 965 hPa, $\tau = 9.1 \text{ s}$) and $D = 2.36 \mu\text{m}$ (JFJ, 645 hPa, $\tau = 6.1 \text{ s}$) assuming an initial particle diameter of $2 \mu\text{m}$. Measurements of a highly hygroscopic aerosol, ammonium nitrate with an initial diameter of $D = 200 \text{ nm}$, show the onset of cloud droplets (no ice crystals since $T < 235 \text{ K}$ for homogeneous freezing) in the $3 \mu\text{m}$ at $S_w = 1.038$, as seen in Figure 1 a), and support the calculated maximum size. Therefore, at $S_w = 1.04$, all particles $\geq 4 \mu\text{m}$ are considered to be ice crystals formed on INPs.

Figure 1 b) shows a measured activation curve of ambient air on the JFJ during a high INP event (7:05 22. March 2020, UTC). The onset of cloud droplets in the $\geq 0.3 \mu\text{m}$ size bin exactly at $S_w = 1$ demonstrates the accuracy of the cloud chamber. At $S_w = 1.043$ the onset of supercooled droplets in the size bin $\geq 1 \mu\text{m}$ is visible. At $S_w = 1.13$ an observed activation fraction of 10^{-3} in the $\geq 3 \mu\text{m}$ -OPC size bin indicates droplets only grow to larger than $3 \mu\text{m}$ at much lower supersaturations. The signal visible in the $\geq 4 \mu\text{m}$ -OPC size bin comes from ice crystals which nucleate at $S_w \geq 1.028$ ($S_i \geq 1.378$), since droplets cannot grow to these sizes but ice crystals can.

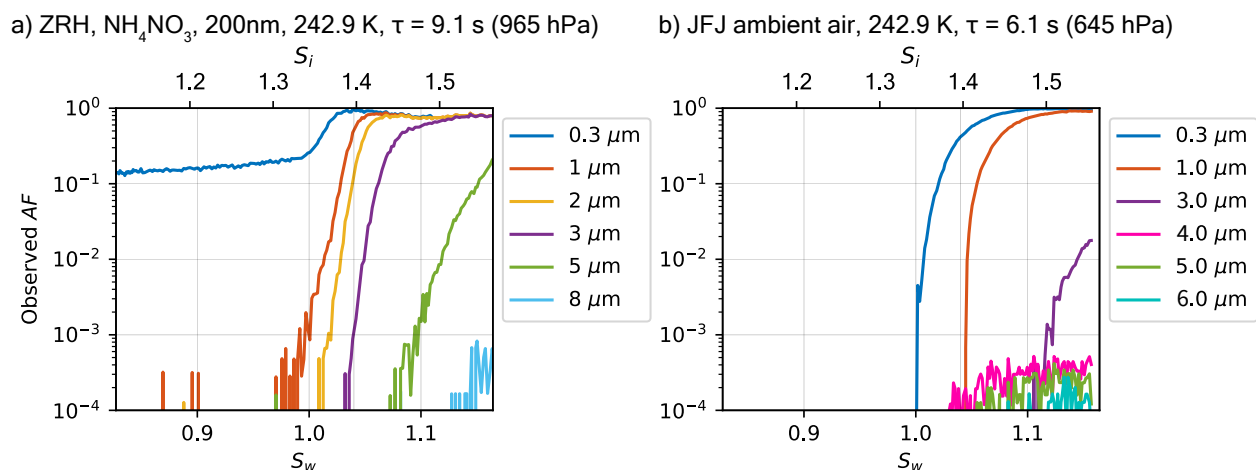


Figure 1: Activation curve at 243 K for a) ambient air, sampled at JFJ during an enhanced INP event, and b) ammonium nitrate, sampled at Zurich. Both measurements were performed under identical flow conditions but resulted in a shorter particle residence time τ at the JFJ compared to Zurich because of the reduced ambient pressure.

False positive counts can arise from large particles other than ice nucleated on an INP. Dominant false positives arise from frost grown on inner chamber surfaces which break off and get carried with the prevailing airflow until exiting the chamber, where they are being detected. To assess and correct the measurements for these particles, after a sampling period of 15 min a background measurement of 5 min is performed. During the background measurement the sample air is guided through a HEPA filter before being sucked into the chamber. The mean time-normalized background counts before and after each INP measurement in the 4

μm bin are subtracted from the $\geq 4 \mu\text{m}$ -OPC counts before the conversion to std L/min. In addition, the limit of detection (LOD) is calculated as followed:

$$LOD = \frac{\sqrt{\sum BG \text{ counts}}}{\sum N_{BG \text{ samples}}} \frac{60}{V t_{OPC}} \quad [LOD] = \text{stdL}/\text{min} \quad (1)$$

Where $\sum BG \text{ counts}$ is the sum of the background counts before and after the measurement, $\sum N_{BG \text{ samples}}$ is the total number of OPC intervals with duration t_{OPC} in seconds used to count all background counts before and after the measurement, and V is the volume flow through the OPC, here $V = 2.83 \text{ std L}/\text{min}$. The stated LOD provides a 62.3% (1σ) confidence interval. The INP concentration falling within the grey bars of the plot are below the LOD. The Minimum Detectable Concentration (MDC, 1 count) for a INP measurement with a sample flow rate of 0.283 std L^{-1} over 15 minutes is 0.236 std L^{-1} and given by the red data points for intervals when a negative concentration was recorded (INP ; BG counts). The green trace is a 2-hour moving average of the concentrations that takes into account all measured INP concentrations (e.g. also negative values). Standard liter (std L^{-1}) is $1\text{e-}3 \text{ m}^3$ at $T = 273 \text{ K}$ and $p = 101325 \text{ Pa}$.

Use of data

Data are available as .CSV files from February 2020 to present at

https://iacweb.ethz.ch/data/hincauto_jfj/

Before using the data for presentations or publications of products resulting from the data, please get in touch with: zamin.kanji@env.ethz.ch

Metadata

Table 1: Content description of the .CSV files found on https://iacweb.ethz.ch/data/hincauto_jfj/

Column	Unit	Description
start(UTC)	UTC	Time stamp at the beginning of the INP measurement. Background sampling is excluded.
end(UTC)	UTC	Time stamp at the end of the INP measurement. Background sampling is excluded.
avgBGcountsBefore	counts 5 s^{-1}	Mean OPC counts during the filtered sampling period (= background) before the INP measurement.
totBGcountsBefore	counts	Total number OPC background counts before the INP measurement.
numOfBGSamplesBefore	5 s^{-1}	Total number of 5 sec OPC background intervals before the INP measurement.
avgINPcounts	counts 5 s^{-1}	Mean OPC counts during the INP sampling period.
avgBGcountsAfter	counts 5 s^{-1}	Mean OPC counts during the filtered sampling period (= background) after the INP measurement.
totBGcountsAfter	counts	Total number OPC background counts after the INP measurement.
numOfBGSamplesAfter	5 s^{-1}	Total number of 5 sec OPC background intervals after the INP measurement.
dedectionLimit	std L^{-1}	Minimum Detectable Concentration, 1 INP count in the INP sample period.
LOD	5 s^{-1}	Limit Of Detection with a 1 σ confidence interval.
LODStdL	std L^{-1}	Limit Of Detection with a 1 σ confidence interval.
INPcorr	counts 5 s^{-1}	INP counts with subtracted mean background counts.
INPcorrStdL	std L^{-1}	Background corrected INP concentration.
TWmean	$^{\circ} \text{C}$	Average warm wall temperature.
TCmean	$^{\circ} \text{C}$	Average cold wall temperature.
Tmean	$^{\circ} \text{C}$	Average center lamina temperature.
Sw		Average supersaturation with respect to water at center lamina.
RHsheath	%	Relative humidity of the sheath air (at Tmean).
RHsample	%	Relative humidity of the sample air after drying (at 293 K).
pSheath	Pa	Pressure within the chamber.
pSample	Pa	Pressure within the sample inlet after drying.
flowSheath	std L min^{-1}	Measured average sheath air flow rate.
flowOPC	std L min^{-1}	Measured average flow rate exiting the chamber.
comment		General comments

std L^{-1} : $1\text{e-}3 \text{ m}^3$ at $T = 273 \text{ K}$ and $p = 101325 \text{ Pa}$.