Temperature and size dependence of volume and surface nucleation rates for homogeneous freezing of supercooled water droplets

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Procedure

- **Experimental**
  - Generate different sizes of ice particles in a flow tube at low (~ 235-240 K) temperatures
  - Measure infrared spectra of the particles during freezing

- **Analysis**
  - Invert spectra to obtain (volume) size distributions
  - Predict volume size distributions with parameterized nucleation/growth model
  - Extract nucleation rates for volume and surface nucleation mechanisms by comparing with measurements
Cryogenic Flowtube - Ice Particle Generation and Measurement

Separate experiments were done with small, medium and large particles injected into a cryogenic flowtube.
Flowtube Conditions from CFD Analysis

Interior baffles improve thermal transfer, control temperature profiles

Calibrated CFD temperatures (error less than ±0.5 K)

Laminar flow; no convective recirculation
Determination of Size Distribution by Inversion of Measured Aerosol Spectrum

\[ E_1, E_2, E_3, \ldots, E_N \cdot P_r = M \]

- Least Squares Fitting of Reference Spectra to the Measured Spectrum

- \( E_r \): Monodisperse extinction spectra for radii \( r \), calculated from measured indices of refraction

- \( P_r \): Size distribution obtained by the fitting procedure

- \( M \): Measured spectrum

Direct Fit: No Assumption about distribution shape.

Procedure:

\[
\min_P \left\{ \left| E \cdot P - M \right|^2 \right\}^{1/2}
\]
Typical Results of Spectral Inversion

- Ice + water spectra calculated from retrieved size distribution, compared with measured spectra.
Determining Surface and Volume Nucleation Rates

- Experimental information available:
  - Temperature and velocity profiles (from CFD)
  - Measured number and size distributions of 240 K water droplets before the experiment and water/ice particles afterwards

![Initial volume distributions of three sizes of water droplets at 240 K](image)
Measured Water and Ice Size Distributions

Temperature dependent volume distributions (medium particles)

Volume fraction frozen

Note shift to larger radius on freezing, due to mass transfer from water to ice
Particle Evolution from Microphysical Model

- Temperature profile calibrated by axial measurements
- Model prediction: nucleation from 10-15 s; growth from 10 to 35 s
- Predicted number and volume distributions of water and ice compared with spectral inversion measurements after 35 s
Microphysical Nucleation/Growth Model\textsuperscript{(1)}

- Separate volume and surface nucleation  \( J_t = J_V V + J_S S \)
  - Classical nucleation formalism  \( J_{V/S}(T) = N_{V/S} \frac{kT}{h} \exp \left( - \frac{\Delta F_{V/S}}{kT} \right) \)
  - Djikaev et al.\textsuperscript{(2)} theory for \( J_S \): germ forms in a thin surface layer

- Nucleation model rates:
  - Parameters: \( A_V, B_V, A_S, B_S \)
  - \( J_V(T) = N_V \frac{kT}{h} \exp \left( - \frac{A_V - B_V T}{kT} \right) \)
  - \( J_S(T) = N_S \frac{kT}{h} \exp \left( - \frac{A_S - B_S T}{kT} \right) \)

- Size distribution binned; includes diffusion-limited mass transfer (condensation and evaporation), Kelvin effect corrections and wall losses

- Vary parameters to fit measured size/volume distributions

\textsuperscript{(1)} Th. Kuhn, \textit{et al.} \textit{ACP} \textbf{11} 1 (2011); M. Earle \textit{et al.} \textit{ACP} \textbf{10}, 7945 (2010)


\textit{ibid} \textbf{112}, 6592-6600 (2008)
Converged Model Predictions vs. Measurements

- Water & ice size distributions and fraction frozen agree well with measurements (~30 experiments with S, M, L particles at various temperatures)

⇒ Model is accurate
Result: Total Nucleation Rate

- Compares well with Stöckel et al. (2005)*; Not well with some others
  - Difficult measurements (nucleation rate changes by nearly a factor of 100 per degree


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Surface and Volume Nucleation Rates Separated

- Surface nucleation rates are higher than volume for these sizes
- Difference is larger for smaller particles

\[ J_V \text{ – solid lines} \]
\[ J_S \text{ – dashed lines} \]
Relative Contributions of Volume and Surface Nucleation

Surface nucleation predominates for $r < 5\mu m$
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Photo: Pekka Parviainen