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# A new method for operating a continuous-flow diffusion chamber to investigate immersion freezing: assessment and performance study

**Gourihar Kulkarni et al.**

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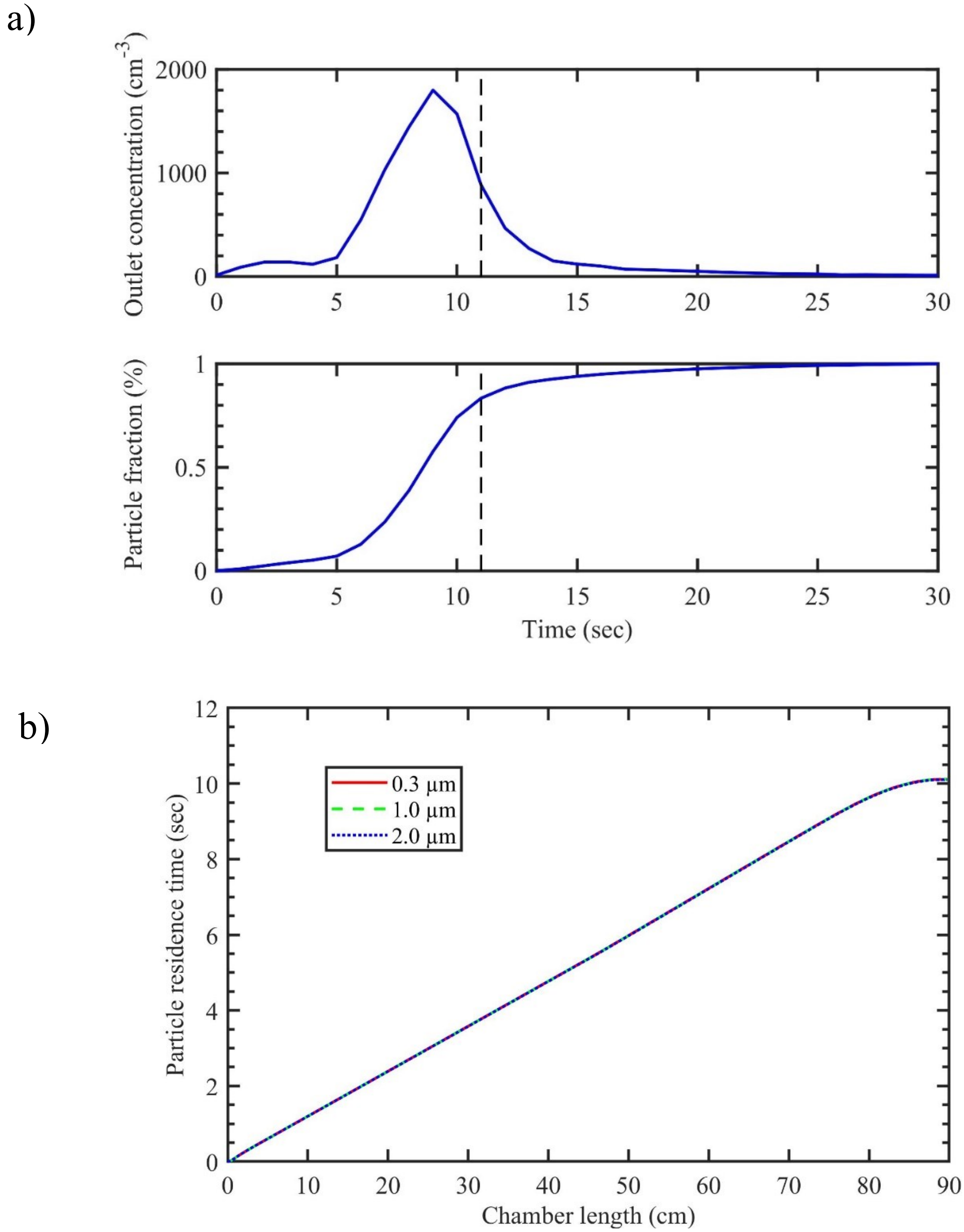
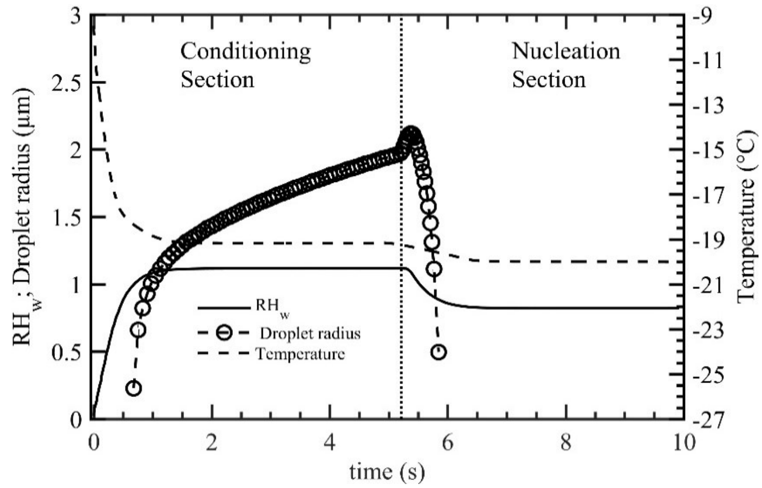
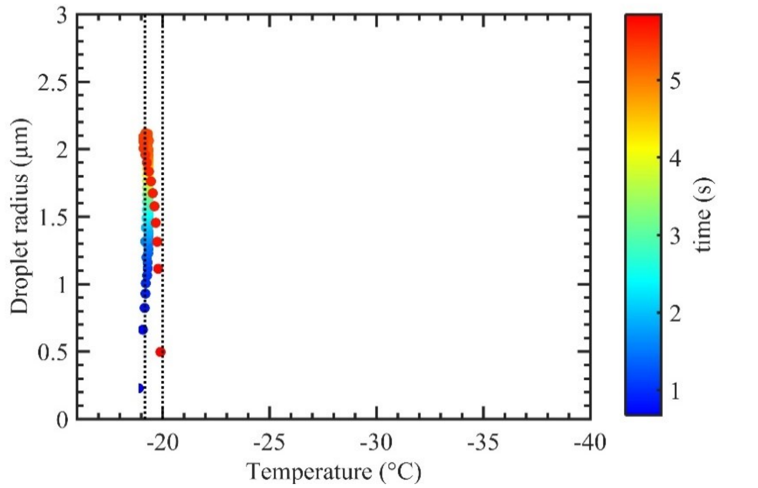



**Figure S1: (a) Measured particle concentration and corresponding particle fraction at the outlet of the ice chamber during particle pulse experiments. The data from three particle pulse experiments were averaged. The vertical dashed line indicates the duration of the particle pulse, and the particles that are measured after this dashed line are classified to be moved out of the aerosol lamina. The data is shown only when CPC started recording the particle counts. (b) Particle residence time of different size droplets within the chamber.**

a)


Conditioning Section
Nucleation Section
RH$_w$; Droplet radius (μm)
Temperature (°C)
time (s)
RH$_w$
Droplet radius
Temperature

b)


Droplet radius (μm)
Temperature (°C)
time (s)

c)

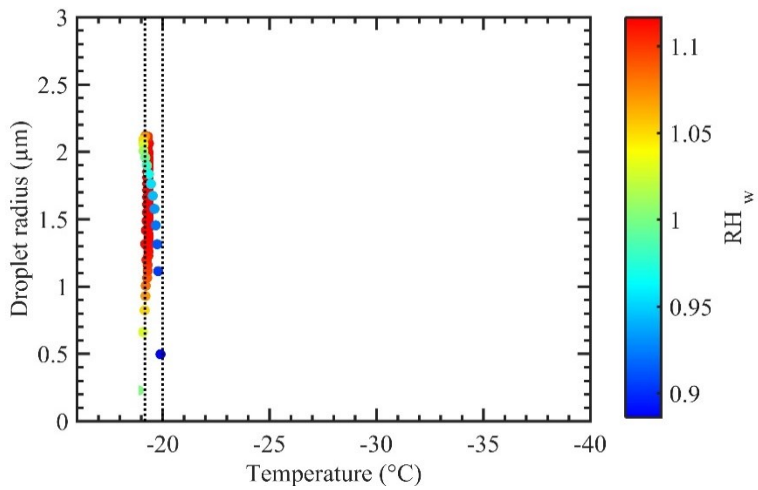

Droplet radius (μm)
Temperature (°C)
RH$_w$

**Figure S2: (a) CFD calculated temperature and *$RH_w$* profiles of a potential INP released from the sample injection region to the outlet end of the chamber. The warm and cold walls of conditioning section are maintained at -9 and -27°C, respectively. The nucleation section is maintained at -20°C. Analytical calculations of droplet growth and evaporation of such a potential INP (0.3 µm in diameter) are also shown. The left and right side of the vertical dotted line represent the conditioning and nucleation sections, respectively. (b) and (c) Alternative representation of similar data shown in panel (a) except as a function of *$RH_w$* and time, respectively. The two vertical dotted lines represent the aerosol lamina temperature (~-19.7°C) and nucleation section temperature (-20°C). Droplets grow for ~5 s to ~4 µm in diameter. Within the nucleation section, these droplets are subjected to colder temperatures than the aerosol lamina temperature and are evaporated.**

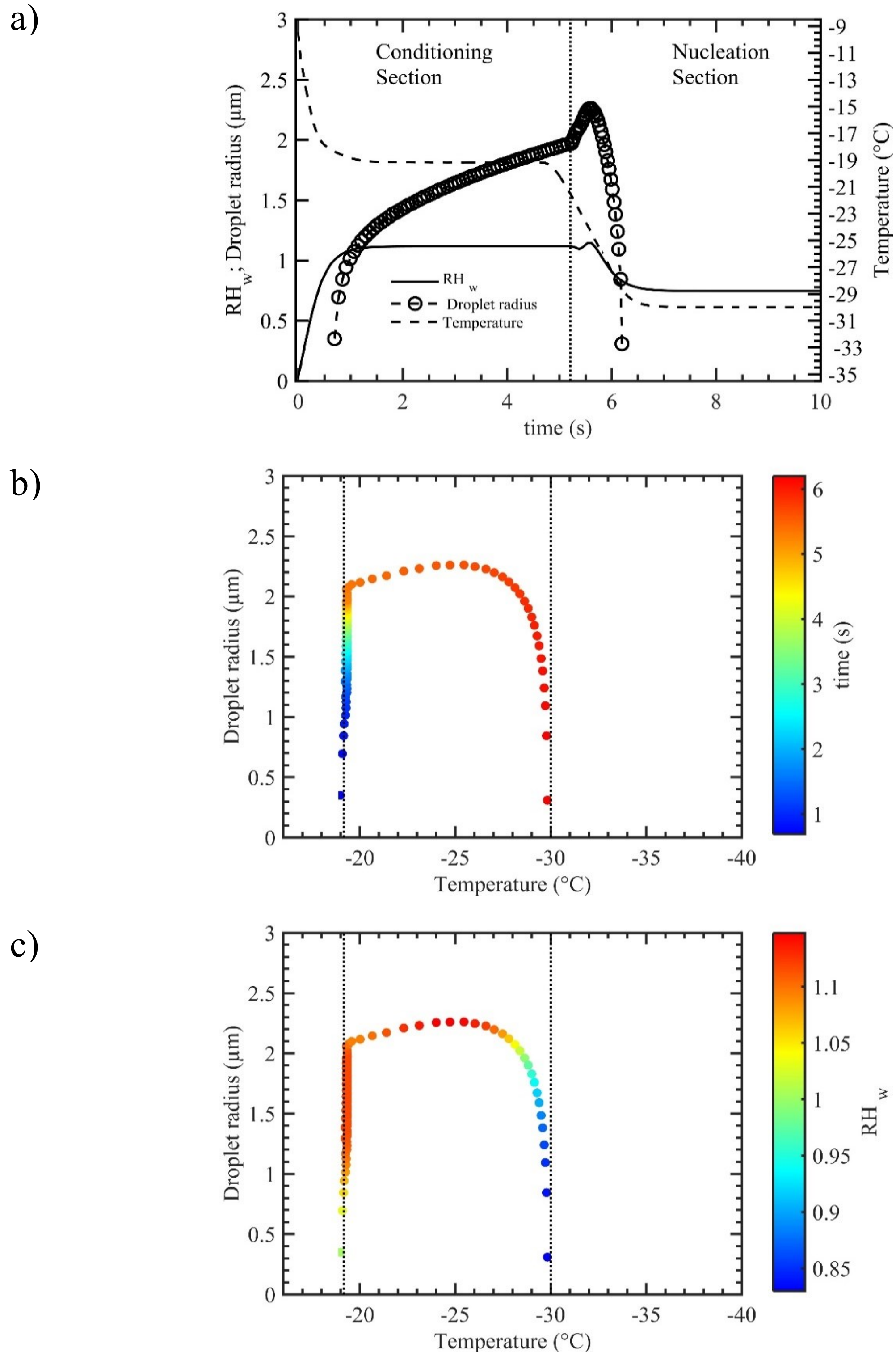



**Figure S3: As in Fig. S2, but nucleation section is maintained at -30°C.**

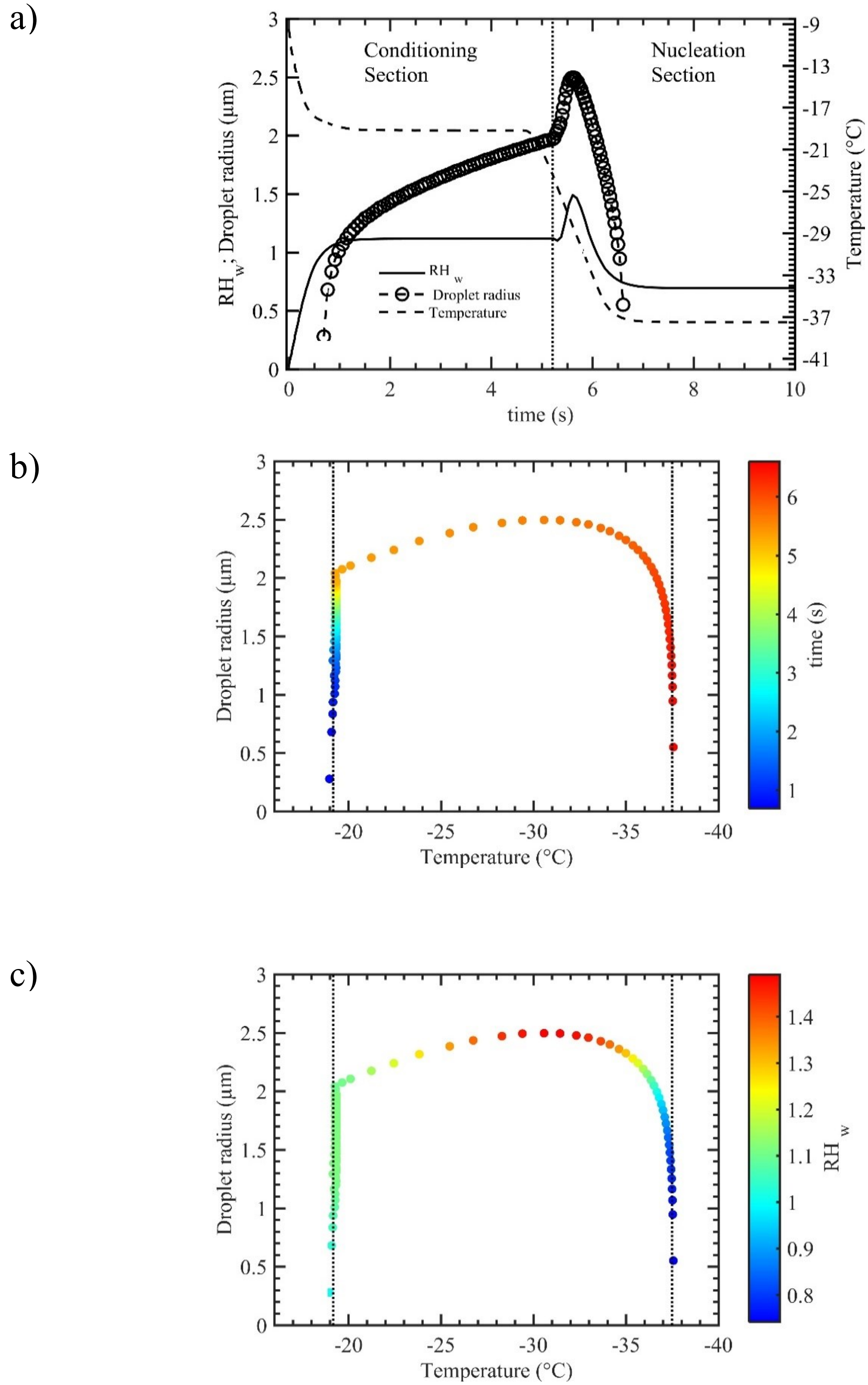



**Figure S4: As in Fig. S2, but nucleation section is maintained at -37.5°C.**

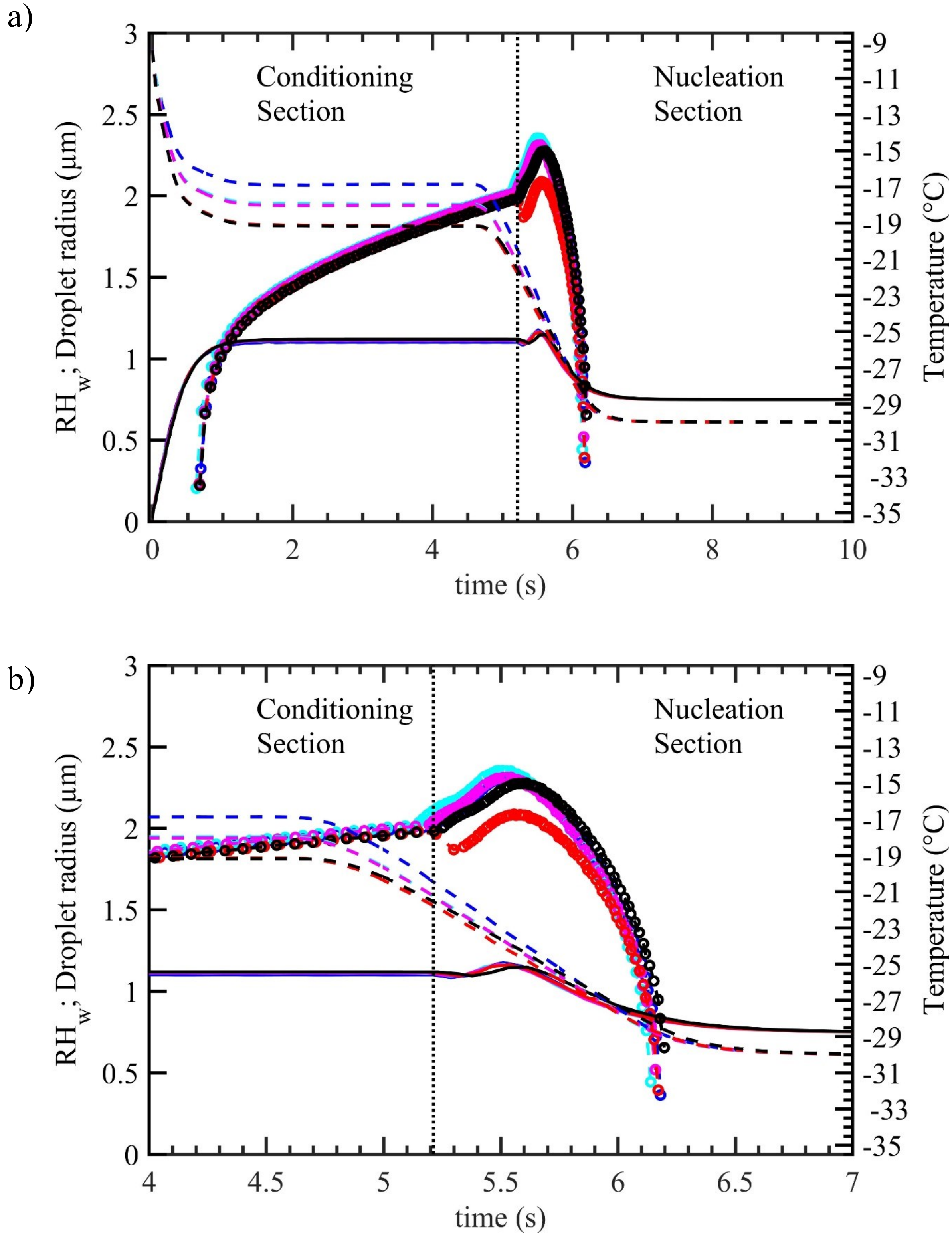
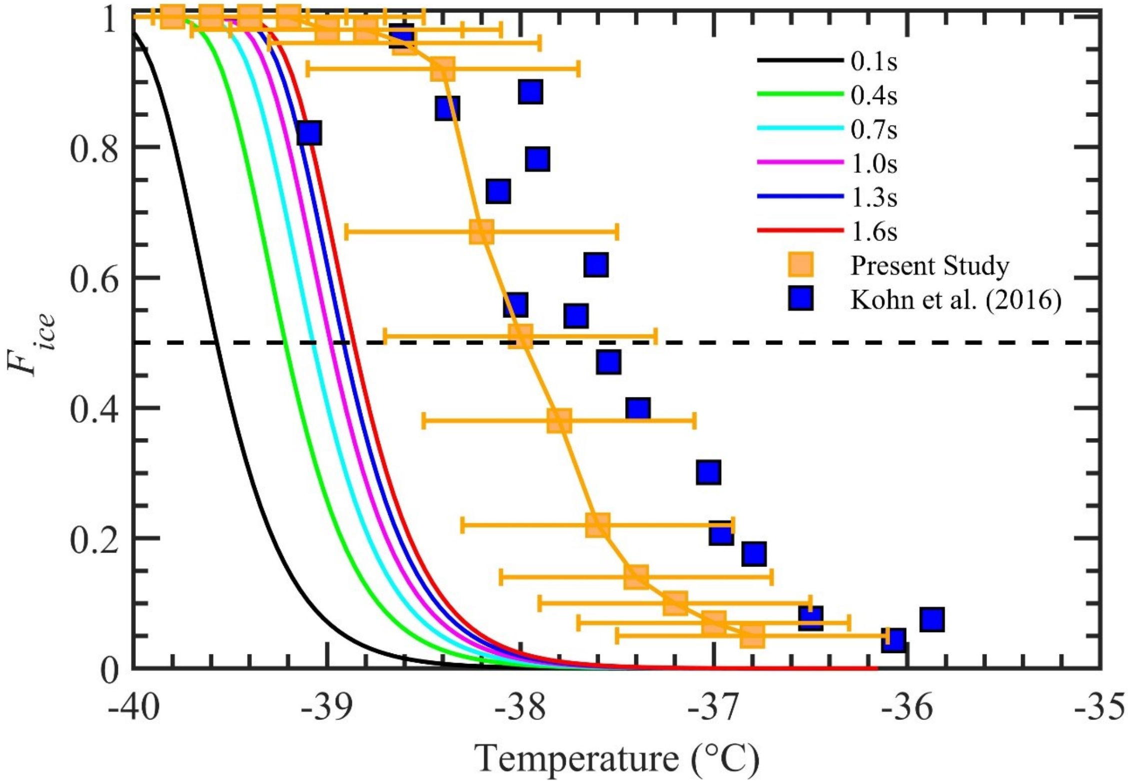


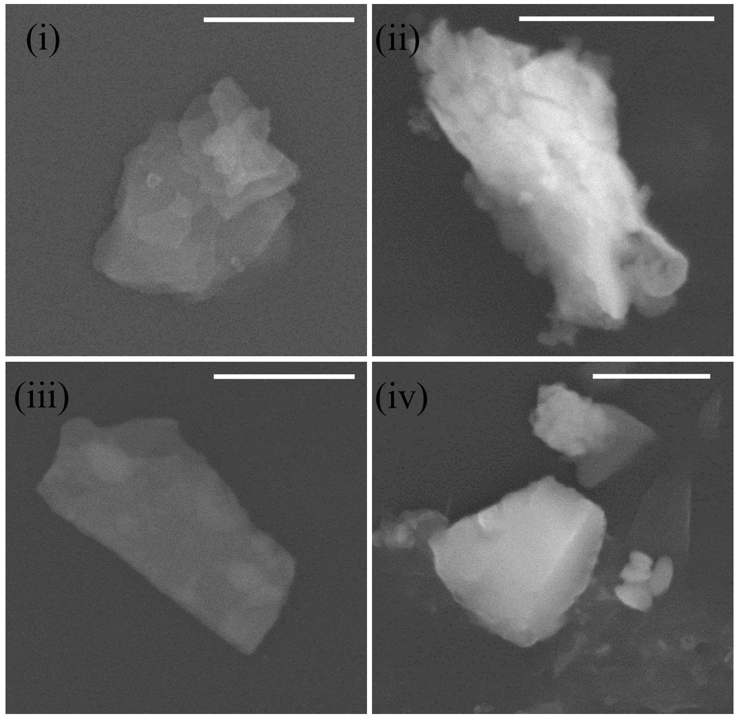

**Figure S5: (a) As in Fig. S3 (a), but data from the five potential INPs. The source positions of these INPs are chosen such that they represent all the particles that are released from the inlet section of the chamber. The black, red, pink, teal, and blue INP trajectories are released 4.35, 4.425, 4.5, 4.575, and 4.65mm distance away from the cold wall (see Figure 3 for more details). The black color trajectory is also shown in Fig. S3 (a). (b) Enlarged view of a junction where both conditioning**

**and nucleation sections meet. The INPs are exposed to range of temperature conditions (-17 to -19.5°C) within the conditioning section but converge to a single temperature after ~0.5s on entering the nucleation section. Such spread in the temperature distribution however does not affect the droplet evaporation kinetics as they all evaporate within ~1s after they enter the nucleation section. The plotted data line style and marker symbol are similar to the legend described in Figure S4.**

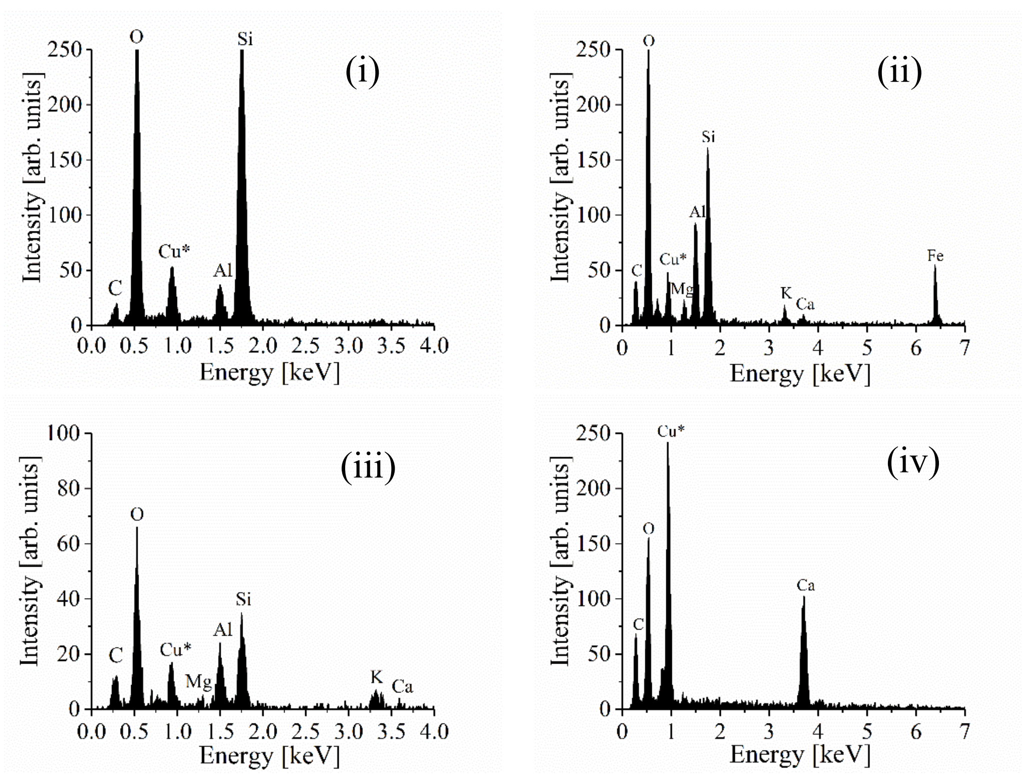



**Figures S6: The theoretical frozen fraction curves of the droplets of diameter 4 µm as a function of probable droplet residence time within the nucleation section. The calculations are based on the homogenous nucleation rate reported previously by other studies, see text for details. Square markers indicate the experimental results from this and previous study (Kohn et al. 2016). Future studies can be employed to extend these calculations to smaller droplets.**

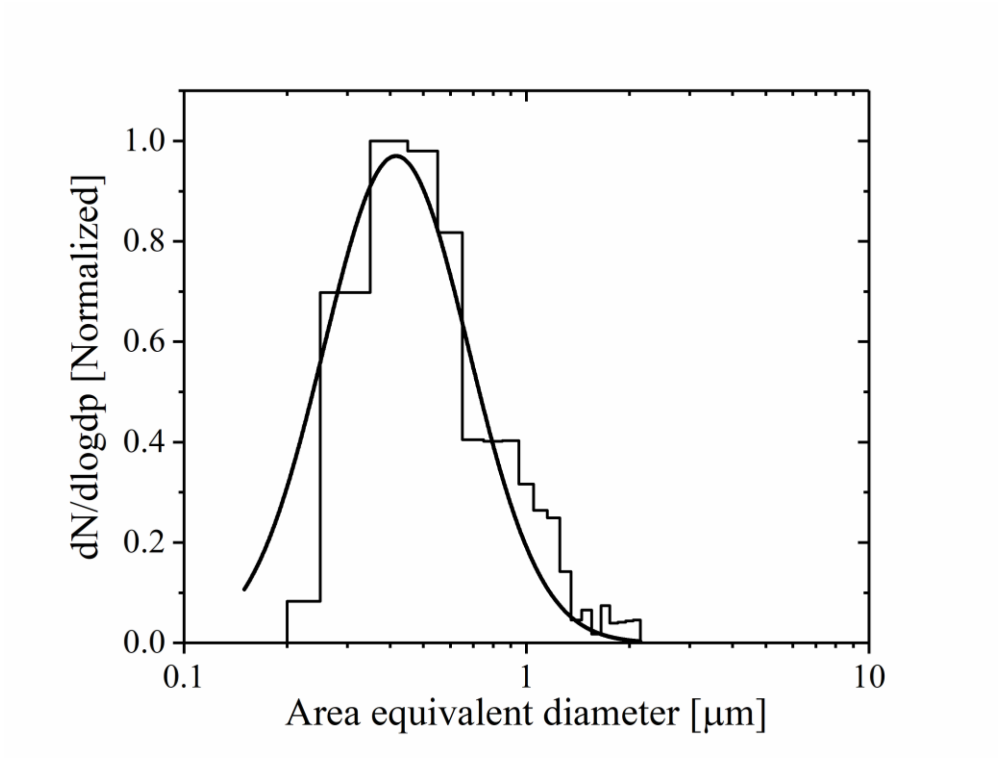
a)


(i)
(ii)
(iii)
(iv)

b)


(i)
(ii)
(iii)
(iv)
Intensity [arb. units]
Energy [keV]
C
O
Cu*
Mg
Al
Si
K
Ca
Fe

c)


dN/dlogdp [Normalized]
1.0
0.8
0.6
0.4
0.2
0.0
0.1
1
10
Area equivalent diameter [µm]

d)

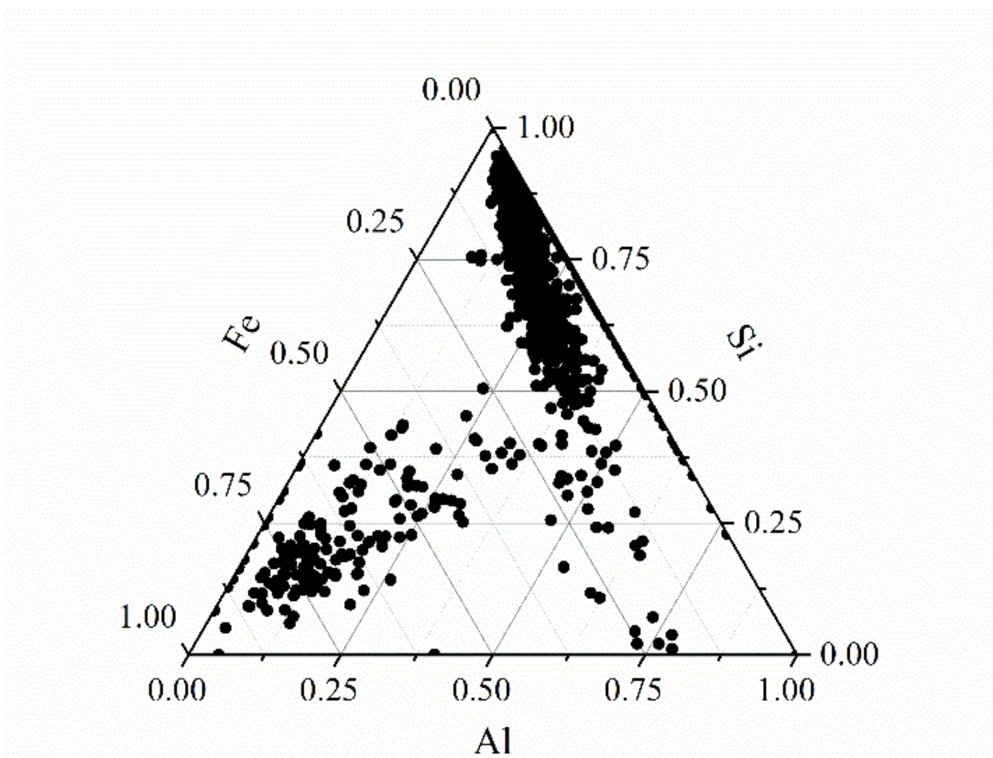

0.00
1.00
0.25
0.75
Fe
0.50
0.50
Si
0.75
0.25
1.00
0.00
0.00
0.25
0.50
0.75
1.00
Al

**Figure S7: Morphology (a), elemental composition (b), size distribution (c), and ternary diagram (d) of airborne desert dust particles sampled at PNNL location. The particle size distribution with a lognormal fit (0.53±0.02 µm) was obtained after analysis of total 1183 particles. Ternary plot of Al, Si, and Fe components of dust particles suggest two distinct types: Si-rich and Fe-rich dust particle. The white bar in (a) represents the scale equal to 1 µm.**

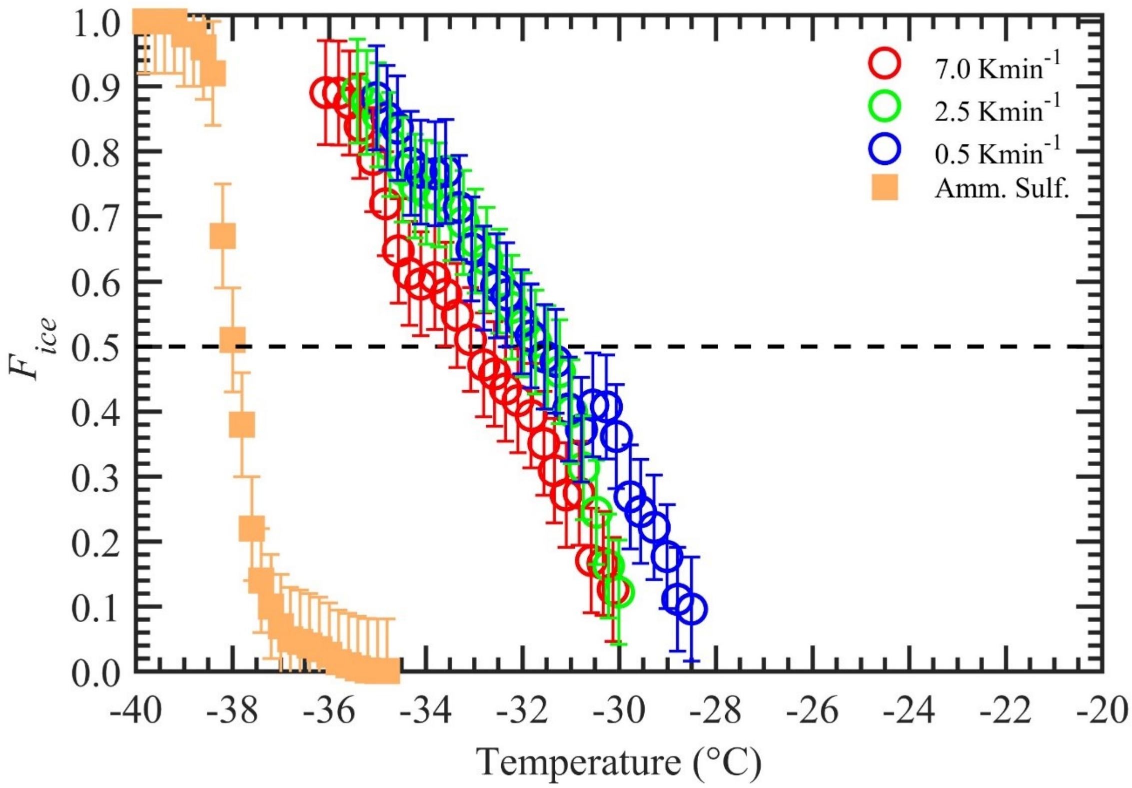



**Figure S8: The $F_{ice}$ of airborne arable dust species as a function of temperature and nucleation section cooling rates. The cooling rate of 0.5 Kmin-1 was used in this study.**

## Text S1 Ice fraction calculations

The ice fraction ($F_{ice}$) as function of temperature ($T$) is calculated as follows:

$$F_{ice}(T) = \frac{N_{ice}(T)}{N_{tot}} \quad (3)$$

where, $N_{ice}(T)$ is the cumulative number of frozen droplets between conditioning section temperature (~-20°C) and set temperature of the nucleation section, and $N_{tot}$ is the total number of droplets that enter the nucleation section. The freezing temperature ($T$) is defined at the steady state temperature of the nucleation section, and the freezing temperature uncertainty is assumed to be similar to the uncertainty across the nucleation section (± 0.4°C).

### Text S2 Droplet growth and evaporation calculations

The droplet growth and evaporation calculations are performed using a water vapor diffusion growth theory (Rogers and Yau, 1989) that neglects temperature corrections and kinetic and ventilation effects as follows:

$$\frac{dm}{dt} = 4\pi r D_v \left(\frac{e_\infty}{R_v T_\infty} - \frac{e_r}{R_v T_r}\right) \quad (1)$$

where

$m$ = mass of the droplet (kg);

$t$ = time (s);

$r$ = radius of the droplet (m);

$D_v$ = diffusion coefficient of water vapor in air (cm$^2$ s$^{-1}$) at far field ambient temperature ($T$ in K) and far field ambient pressure ($p$ in mb) is given by,

$$D_v = 0.211 \left(\frac{T}{273.15}\right)^{1.94} \left(\frac{1013.25}{p}\right)$$

$e_\infty$ and $e_r$ = far field ambient and droplet surface water vapor pressure (Pa), respectively;

$R_v$ = gas constant for a unit mass of water vapor (J K$^{-1}$ kg$^{-1}$);

$T_\infty$ and $T_r$ = far field ambient and droplet surface temperature (K), respectively.

Equation (1) can be solved to obtain the final radius ($r_1$ in m) of the droplet as follows:

$$r_1 = \left(\frac{3}{\rho}\, r_0 D_v \left(\frac{e_\infty}{R_v T_\infty} - \frac{e_r}{R_v T_r}\right) t + r_0^3\right)^{1/3} \quad (2)$$

where $r_0$ is the initial radius of the droplet. Equation (2) was used to calculate the droplet growth and evaporation.

**Table S1: The evaporative cooling of an individual droplet of 4 µm in diameter cooled at different temperature and humidity conditions. The maximum supercooling of the droplet (=0.26 °C) is observed at -20 °C and a saturation ratio of 0.8. The cooling is very small (<0.001) for certain temperatures at $RH_w$ = 0.99.**

| Ambient Temperature (°C) | Evaporative cooling (°C) Ambient saturation ratio | | |
|---|---|---|---|
| | *0.99* | *0.9* | *0.8* |
| *-20* | 0.01 | 0.12 | 0.26 |
| *-30* | < 0.001 | 0.05 | 0.11 |
| *-40* | < 0.001 | 0.02 | 0.04 |