

**Supplementary Information**

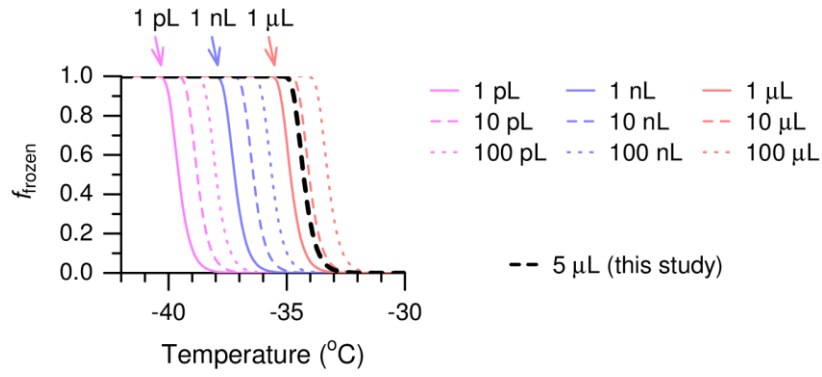
**An improved approach for measuring immersion freezing in large droplets over a wide temperature range**

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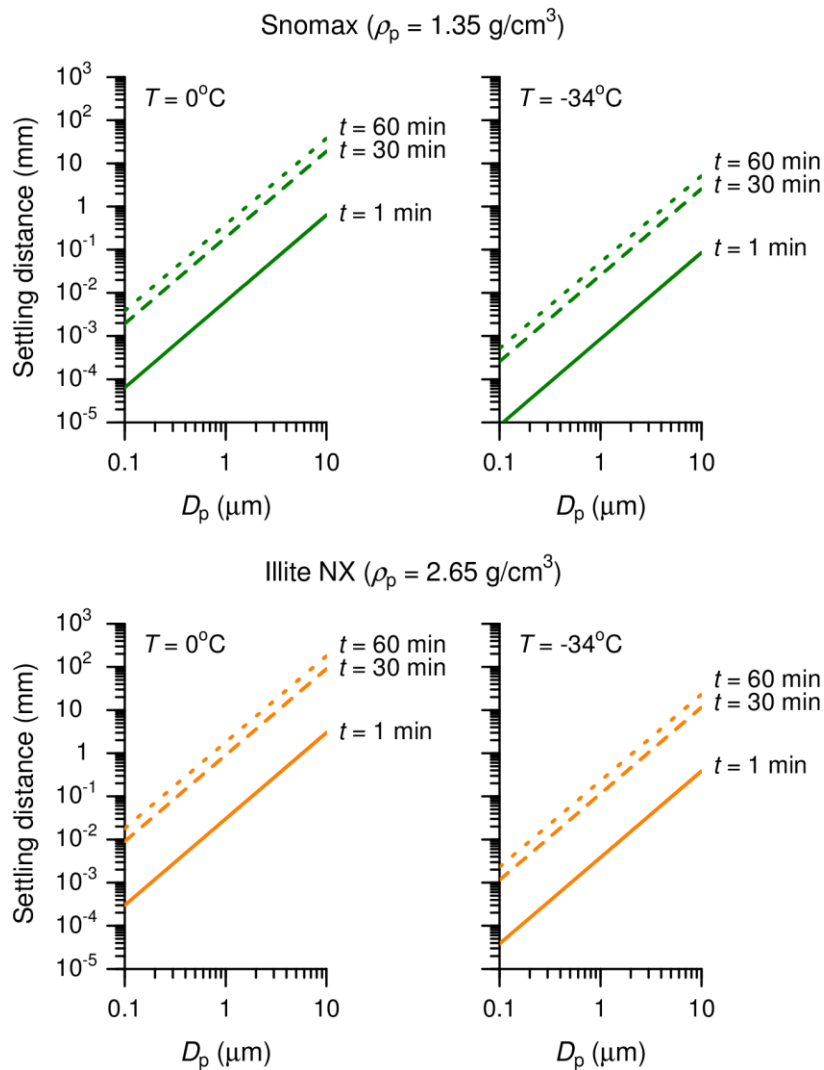
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**Supplementary Figure S1. Fraction of droplets frozen as a function of temperature for pure water droplets of different volumes.** These values are obtained using a classical nucleation theory-based parameterization for homogeneous freezing with a cooling rate of  $1^\circ\text{C}/\text{min}$  (ref. 32).



**Supplementary Figure S2. Size dependence of the settling distances of Snomax or illite NX particles in water at temperatures of  $0^\circ\text{C}$  and  $-34^\circ\text{C}$ .** The settling distances after 1, 30 and 60 minutes are estimated based on the terminal setting velocities of the particles (see Supplementary Methods).

## Supplementary Methods

According to Stokes' law, the terminal settling velocity ( $v$ ) of small spherical particles in water is expressed as:

$$v = \frac{(\rho_p - \rho_w) \cdot g}{18\eta_w} \cdot D_p^2 \quad (S1)$$

where  $\eta_w$  is the viscosity of water,  $\rho_w$  is the density of water,  $\rho_p$  is the density of particles,  $D_p$  is the particle diameter, and  $g$  is the gravitational acceleration ( $= 9.80665 \text{ m/s}^2$ ). In the calculations, it is assumed that the  $\eta_w$  and  $\rho_w$  values are temperature dependent parameters in the temperature range between  $-34^\circ\text{C}$  and  $0^\circ\text{C}$  (refs 37 and 38) and that the  $\rho_p$  values of Snomax and illite NX particles are  $\sim 1.35 \text{ g/cm}^3$  and  $\sim 2.65 \text{ g/cm}^3$ , respectively<sup>29,30</sup>. The settling distances of Snomax and illite NX particles in water at  $0^\circ\text{C}$  and  $-34^\circ\text{C}$  are then estimated using the  $v$  values. Supplementary Figure S2 illustrates the settling distances after  $t$  minutes ( $t = 1, 30$  and  $60$ ). Given that the diameters of Snomax and illite NX particles used here are mainly distributed between about  $0.1$  and  $1 \text{ }\mu\text{m}$  (refs 29 and 30), it is expected that the majority of them are suspended in  $5 \text{ }\mu\text{L}$  water droplets (a few micrometers in diameter) even at the end of each freezing experiment (i.e., the settling distances of the particles are shorter than the diameter of the droplets).

## References

37. Dehaoui, A., Issenmann, B. & Caupin, F. Viscosity of deeply supercooled water and its coupling to molecular diffusion. *Proc. Natl Acad. Soc. USA* **112**, 12020-12025 (2015).
38. Hare, D. E. & Sorensen, C. M. The density of supercooled water. II. bulk samples cooled to the homogeneous nucleation limit. *J. Chem. Phys.* **87**, 4840-4845 (1987).