Immersion freezing by Snomax™ particles: Comparison of results from different instruments

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Introduction

- ice nucleation, the first step in the generation of ice in clouds, still poses open questions
- heterogenous ice nucleation processes, among them immersion freezing, are particularly important for ice fomation in mixed phase clouds
- there are still large discrepancies in results obtained for immersion freezing when different measurement methods / instrumentations are compared (Hoose & Möhler (2012), Murray et al. (2012))
- within the DFG funded research unit INUIT (Ice Nucleation research UnIT, FOR 1525) we made an effort to compare immersion freezing measured by a suite of different methods / instruments
- in this poster, results for the comparison of immersion freezing induced by Snomax™ are presented
- Snomax™ is commercially available (used in artificial snow production), containing non-viable *Pseudomonas syringae* bacteria (known to be ice active) and has been used as model substance for *P. syringae* in the past
- Snomax™ from the same badge was distributed to all participating groups
- likewise, the same atomizer was used by all groups when possible; some of the measurements were conducted during a measurement campaign at LACIS (Leipzig Aerosol Cloud Interaction Simulator)
- the following instruments participated in the comparison: an acoustic levitator (AcLev, Diehl et al., 2009), AIDA (Hiranuma et al., 2014), BINARY (Budke et al., 2013), FINCH (Bundke et al., 2008), LACIS (Hartmann et al., 2011), PINC (Chou et al., 2011) and the Mainz vertical windtunnel (WT, Diehl et al., 2011)

Particle generation

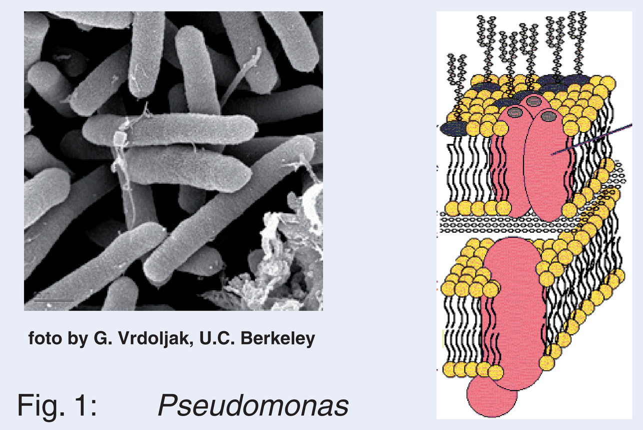
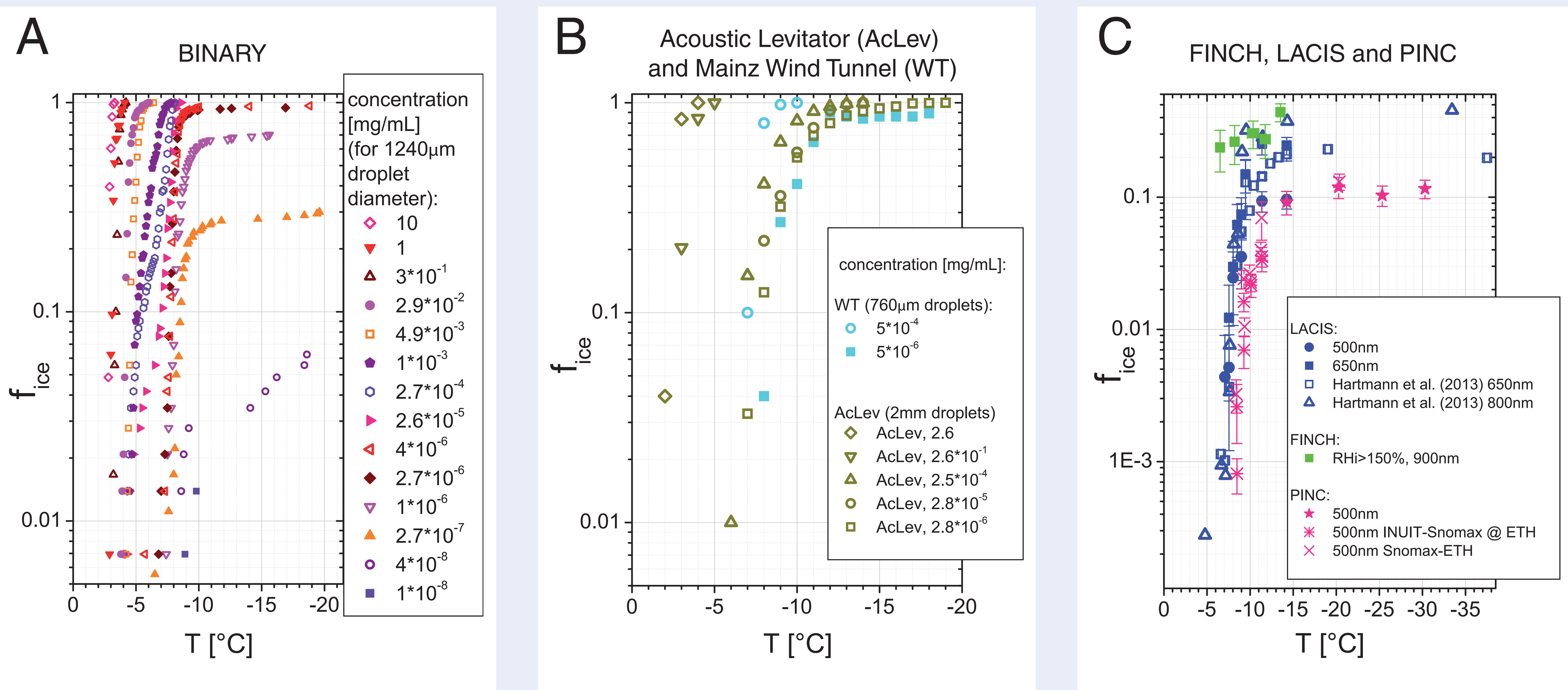


Fig. 1: *Pseudomonas syringae* belong to those bacteria that can form ice nucleation active (INA) protein complexes (anchored in the bacterial membrane).

- Snomax™ suspensions and Snomax™ particles may contain: whole bacteria cells and fragments of cell membrane which both might carry an ice nucleation active (INA) protein complex (known to induce the ice nucleation), remnants of the nutrients the bacteria were grown in, and material leeching from the inner parts of the bacteria
- some data shown in this poster were obtained by examining the freezing of droplets prepared from Snomax™ suspensions of various concentrations, spanning a range of 10 orders of magnitude in concentration (AcLev, BINARY, WT)
- some methods used Snomax™ suspension to generate dry particles, which were then activated to droplets in the set-up and cooled subsequently (AIDA, FINCH, LACIS, PINC); this was done as follows:
  - spraying of an aqueous suspension from Snomax™ and subsequent drying
  - size selection using a diffusion dryer and a Differential Mobility Analyzer (DMA); dry sizes were selected in a size range from 200nm to 900nm

Measured frozen fractions



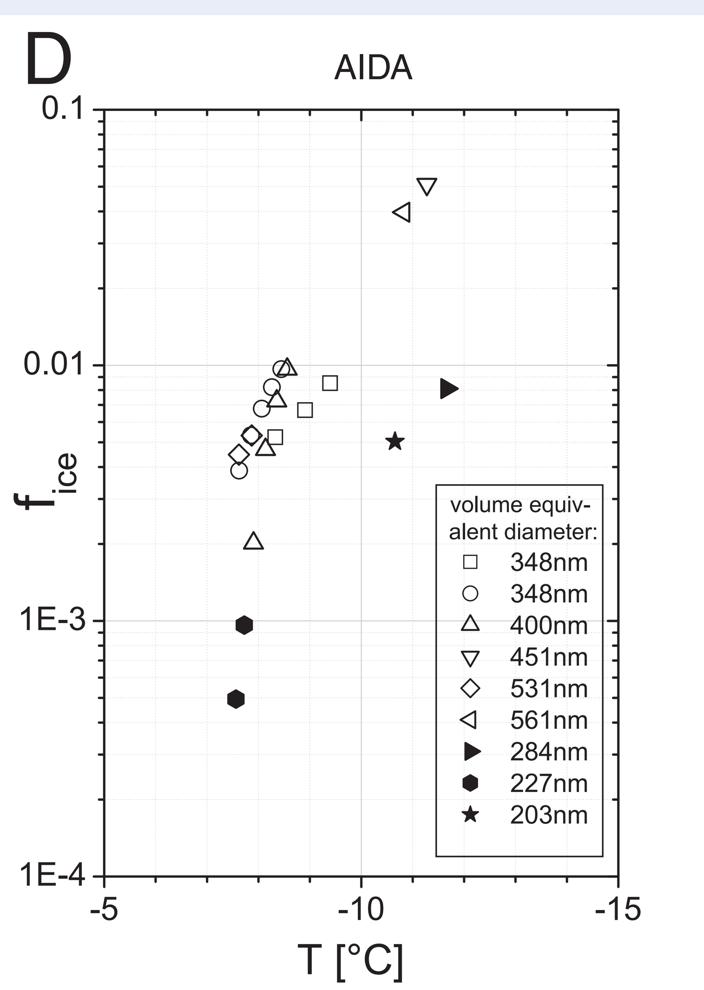


Fig. 2 (A-D): Frozen fractions (f_{ice}) as measured by the different methods / instruments (for LACIS and PINC (C, i.e. upper right panel), data for particles generated from a different Snomax™ badge while using a different atomizer are included (labeled "Hartmann et al. (2013)" and "ETH"); for AIDA (D, lower panel), open symbols correspond to measurements for which monodisperse particles were selected with a DMA, filled symbols represent measurements for which a polydisperse aerosol was used (volume equivalent diameters are given))

- in general, there is a sharp increase in f_{ice} at T < -10°C; if concentrations / particle sizes are so low that not all particles contain an INA protein complex, a plateau forms at f_{ice} < 1
- both, larger droplet concentrations and larger particle diameters correspond to a higher average number of INA protein complexes per droplet / particle, leading to a shift of the freezing curves towards higher temperatures (see also Hartmann et al., 2013)

Theoretical Background

to enable a comparison of the different data-sets, we used a data evaluation based on a description suggested by Vali (1971) and again by Murray et al. (2012) for immersion freezing of droplets containing biological material:

$$f_{ice}(T) = 1 - \exp(-n_m(T)\, C_m\, V) \qquad (1)$$

$n_m(T)$ is the number of INA protein complexes per unit of dry Snomax™ mass, C_m is the concentration of Snomax™ in the examined suspensions and V is the droplet volume

for methods based on the examination of dry particles, the total mass of Snomax™ was calculated assuming spherical particles and a density (ρ) of 1.35 g/cm³:

$$f_{ice}(T) = 1 - \exp(-n_m(T)\, \rho\, \pi/6\, d_p^3) \qquad (2)$$

where d_p^3 is the diameter of the dry Snomax™ particles

n_m is shown in Fig. 3; a value was derived for each of the data points shown in Fig. 2

Active Site Density per Snomax™ Mass

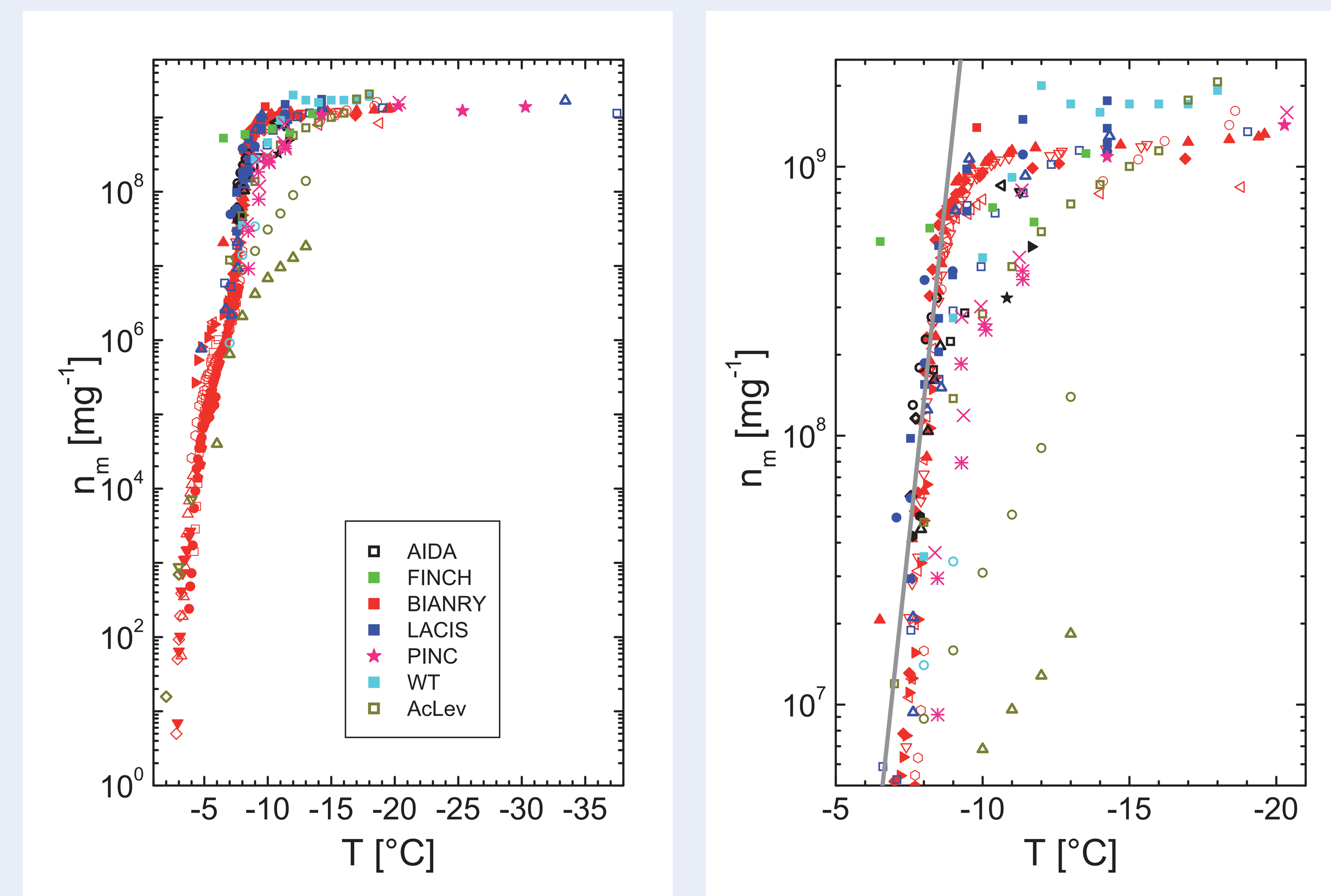


Fig. 3: n_m, i.e. the number of INA protein complexes per mass of Snomax™, is shown in both panels (panel on the right shows an enlargement). Values were derived from f_{ice} shown in Figs. 2 (A-D), using Eqs. (1) and (2). The same symbols were used as in Figs. 2, besides for BINARY data which are all displayed in red, here.

- n_m from most methods / instruments fall together; exceptions are AcLev (the majority of the values is clearly lower than the bulk of data) and FINCH at the highest temperature (it is the only instrument detecting a considerable amount of ice at -6.5°C)
- in Snomax™, two types of INA protein complexes are present, a larger one which becomes ice active already at T > -7°C (see shoulder clearly visible in the BINARY data for $n_m < 2*10^6$ mg⁻¹) and a smaller one which occurs roughly 1000 times more often, being ice active at T < -7°C (for details on the two types see Hartmann et al. (2013) and references therein)
- for T > -10°C, a steep increase of n_m with decreasing T is visible, as more and more of the INA protein complexes become ice active (same slope as found for nucleation rates of the smaller Snomax™ INA protein complex in Hartmann et al. (2013) for roughly -7°C > T > -9°C, see grey curve in right panel (~exp(-2.34*T)))
- at T < -12°C: n_m levels off, forming a plateau, which yields the number of INA protein complexes which are generally present per mass of Snomax™ (1.4*10⁹ mg⁻¹)

Summary

- the immersion freezing of Snomax™ measured by a suite of different methods / instruments could be represented using an active site density per Snomax™ mass, n_m (where the term "active sites" represents the INA protein complexes); this approach assumes the ice nucleation to be deterministic (time independent), consistent with the steep increase in n_m
- values of n_m mostly agree within an order of magnitude, although the different methods covered a wide range of examined droplet concentrations / particle sizes (10 orders of magnitude) and a wide range in ice nucleation times / cooling rates (LACIS being the fastest with an ice nucleation time < 1s, while WT data were recorded after 30s and BINARY had a cooling rate of 1K/min)

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