Karlsruhe Institute of Technology

Micro2Macro
Origins of Climate Change Uncertainty
Image credit: Jeremy Young and Gabor Vali

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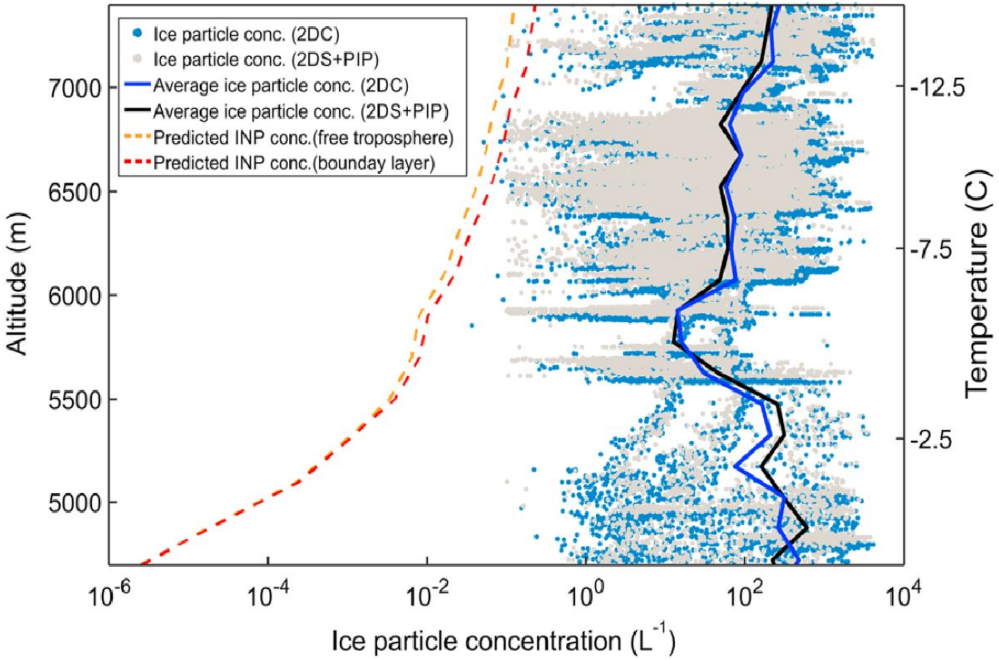
TROPOS
Leibniz Institute for Tropospheric Research

Ice multiplication as a long-standing questi in cloud microphysics

Alexei Kiselev, Alice Keinert, Susann Hartmann, Johanna Seidel, and Thomas Leisner

How Can We Plan Future Process Observations to Reduce Climate Change Uncertainty?

Cloud ice mysteries



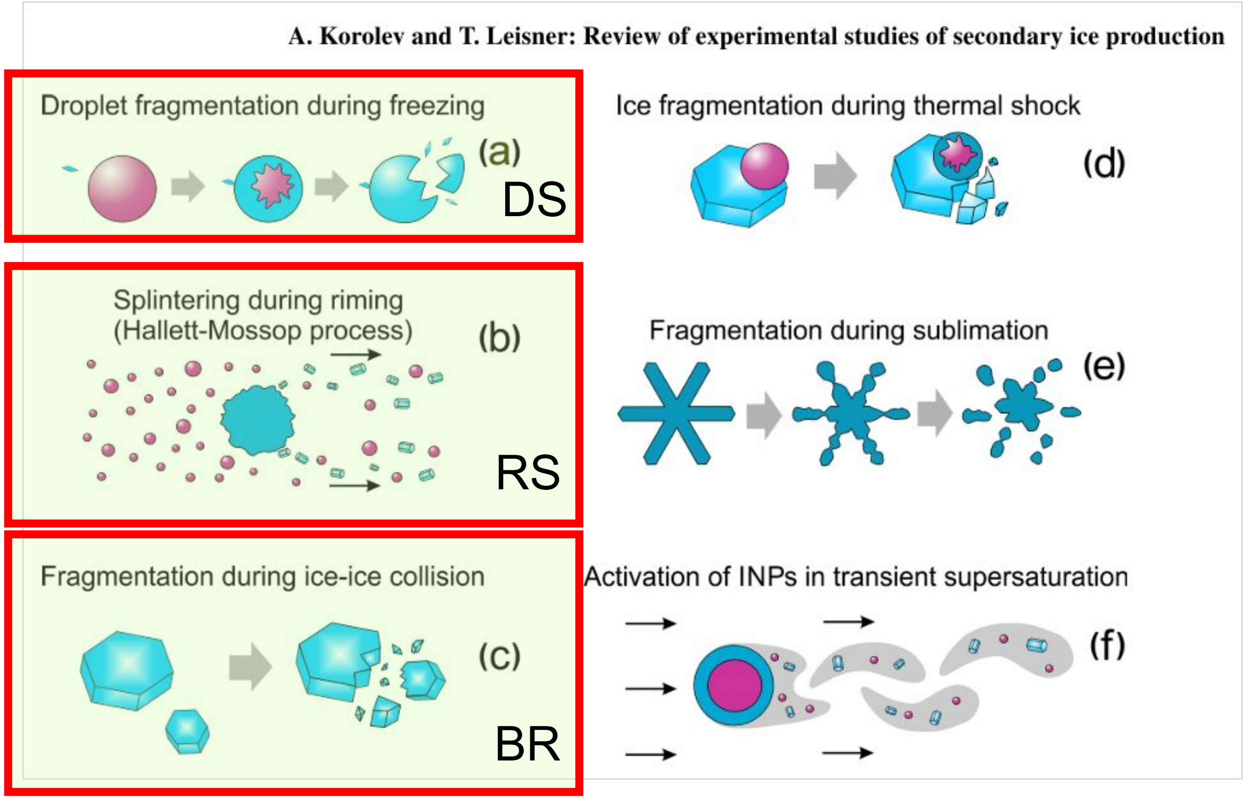
Comparisons of measured average concentrations of INP (dashed lines) and ice crystals (solid lines). INP number concentration obtained via DeMott et al. (PNAS, 2010) parameterization from the measurements of aerosol concentration. Ladino et al. (GRL, 2017)

- Rapid glaciation of convective clouds
- Observation of cloud regions with “explosive” ice particle concentration
- Outstanding inconsistency between INP concentration and concentration of ice particles

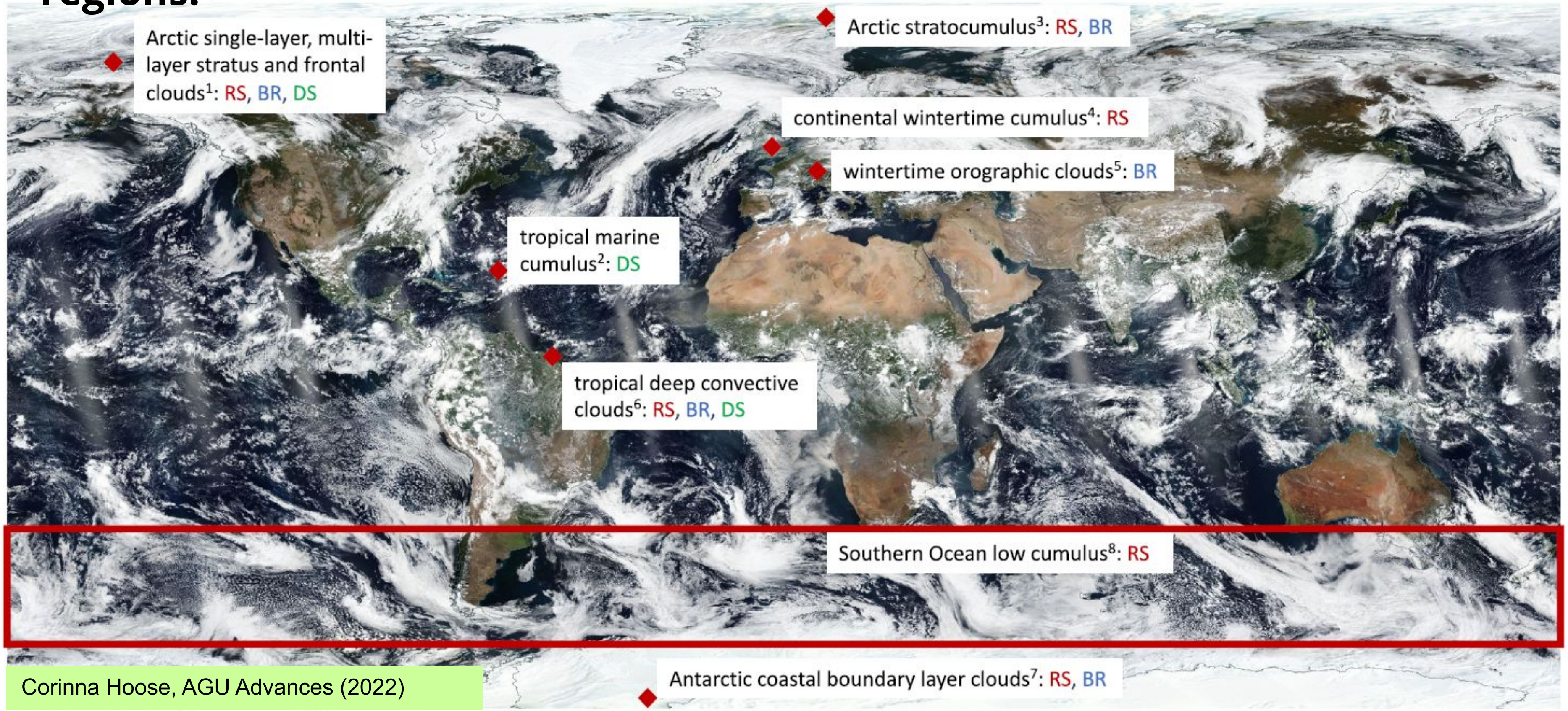
Possible explanations:

- Compromised INP measurements
- Compromised ice particle measurements
- **Formation of atmospheric ice as a result of processes involving pre-existing ice particles: secondary ice production (SIP)**

Hints on possible SIP mechanisms originated in the early lab studies



Ice multiplication is active across different cloud types and regions.



190+

SIP-related studies published in the last 10 years

9

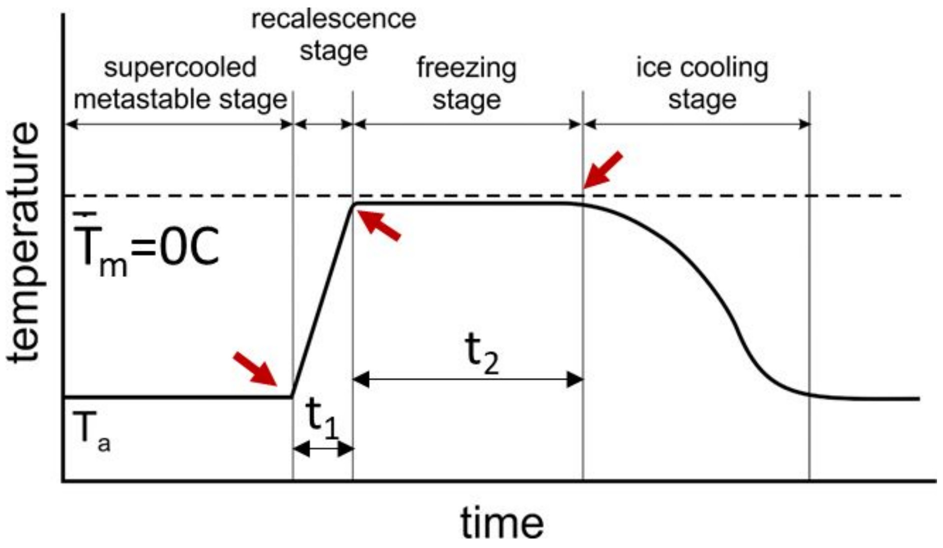
Lab studies

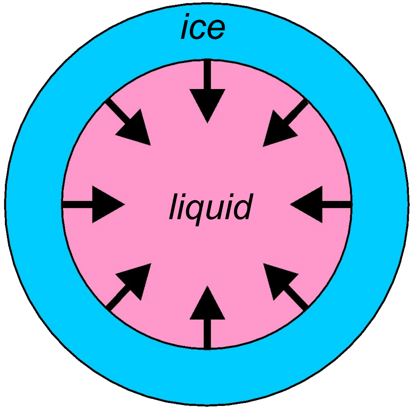
1. **Seidel, J., et al., Secondary ice production – no evidence of efficient rime-splintering mechanism, ACP, 2024.**
2. Grzegorczyk, P. et al., Fragmentation of ice particles: laboratory experiments on graupel–graupel and graupel–snowflake collisions, ACP, 2023.
3. James, R. L., Phillips, V. T. J., and Connolly, P. J.: Secondary ice production during the break-up of freezing water drops on impact with ice particles, ACP, 2021.
4. **Kleinheins, et al.: Thermal imaging of freezing drizzle droplets: pressure release events as a source of secondary ice particles, JAS, 2021.**
5. **Keinert et al. : Secondary Ice Production upon Freezing of Freely Falling Drizzle Droplets, JAS, 2020.**
6. Prabhakaran et al.: High Supersaturation in the Wake of Falling Hydrometeors: Implications for Cloud Invigoration and Ice Nucleation, GRL, 2020.
7. **Lauber et al.: Secondary Ice Formation during Freezing of Levitated Droplet, JAS, 2018.**
8. Emersic, C. and Connolly, P. J.: Microscopic observations of riming on an ice surface using high speed video, Atmospheric Research, 2017.
9. Wildeman et al.: Fast Dynamics of Water Droplets Freezing from the Outside In, PRL, 2017.

Lab studies of secondary ice are challenging

Lab studies example 1. Fragmentation during droplet freezing

Early lab experiments: Visage (1968), King and Fletcher (1973)





$$1\mu s < t_1 < 1s$$
$$1s < t_2 < 10^2 s$$
$$\Rightarrow \quad t_2 \gg t_1$$

By the end of the recalescence stage at -40°C approximately 60% of water remains in liquid phase

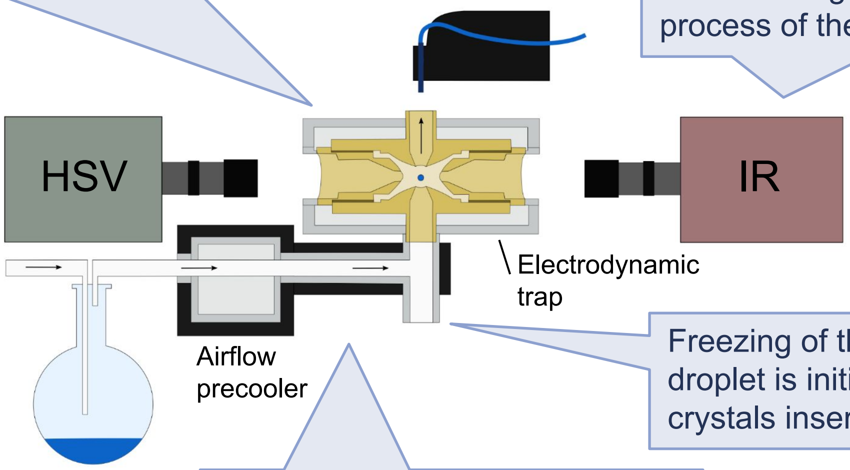
Slide provided by Alexei Korolev

Experimental setup at KIT

Individual water droplets (~300 µm in diameter) are levitated in a temperature controlled electrodynamic balance setup.

Droplet freezing and SIP events are observed with a high speed video (**HSV**) camera (Phantom v710, Vision Research).

The recently added infrared (**IR**) camera (InfraTec) allows for further detailed insight into the freezing process of the droplet.



Freezing of the supercooled droplet is initiated by fine ice crystals inserted into the airflow.

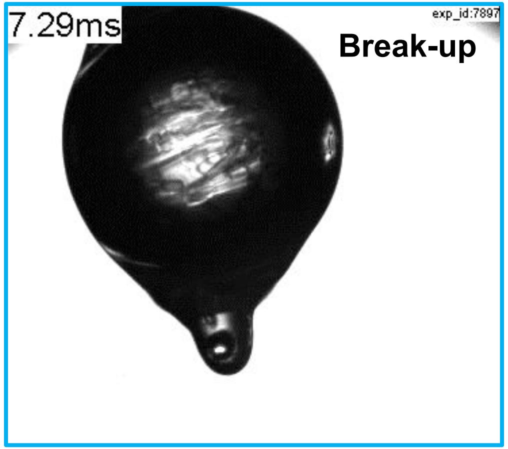
Droplets are exposed to a flow of cold moist air from below, simulating free fall conditions.

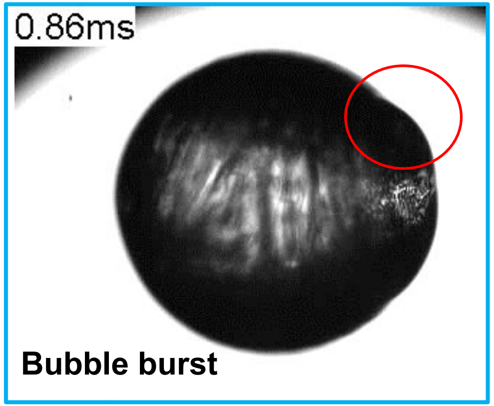
Experimental conditions:

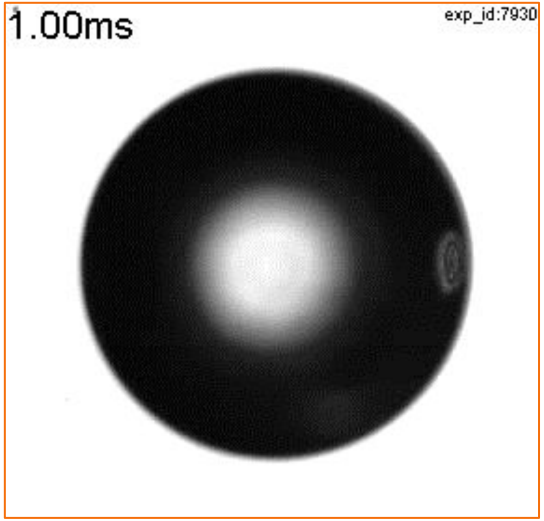
Pure water droplets and droplets of aqueous solution of sea salt analogue (2.9 mg/L SSA)

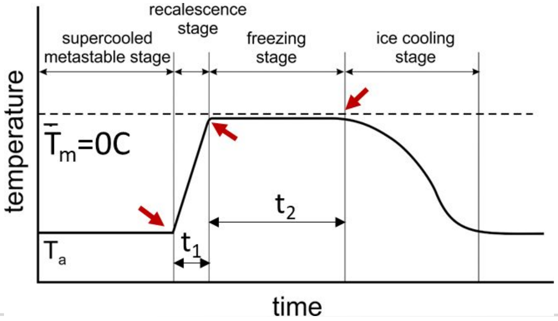
Airflow temperature between -1°C and -30 °C

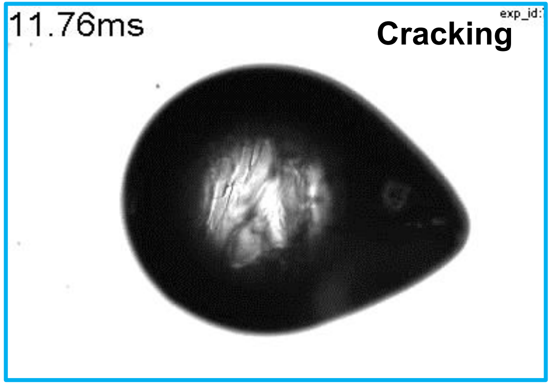
Secondary Ice Production upon freezing of drizzle droplets

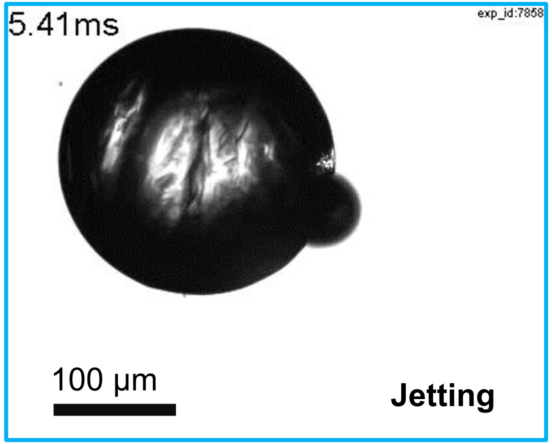












Enhancement of secondary ice production for droplets freezing in free fall

ventilation

droplet rotation

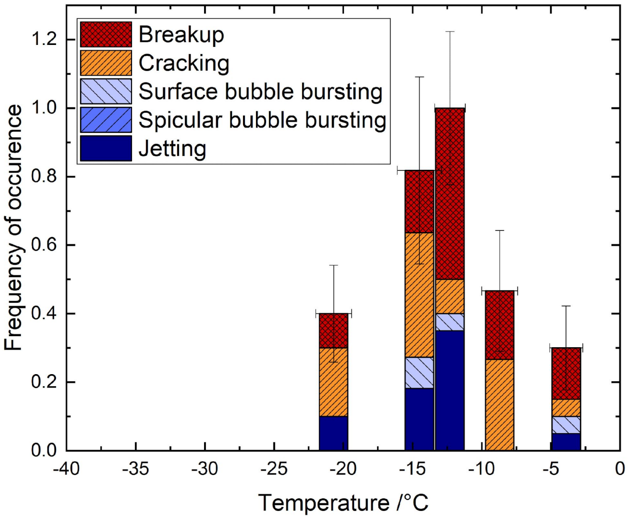
fast symmetrical removal of latent heat

faster ice growth

More potential SIP events

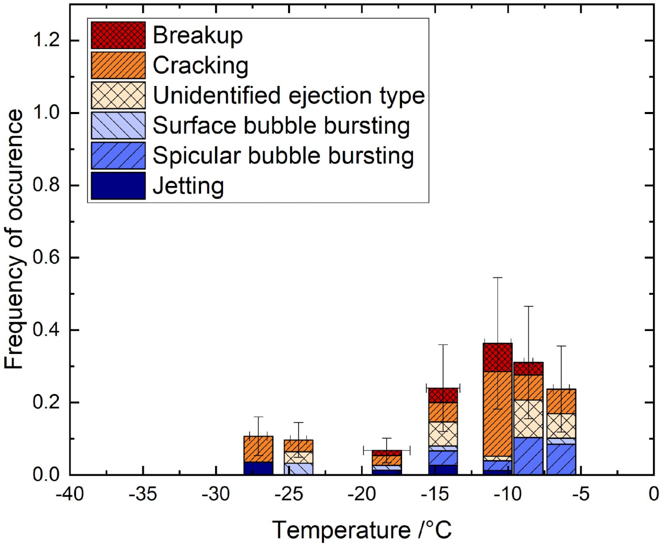
Moist airflow

**(Keinert et al., 2020, JAS
Kleinheins et al., 2021)**

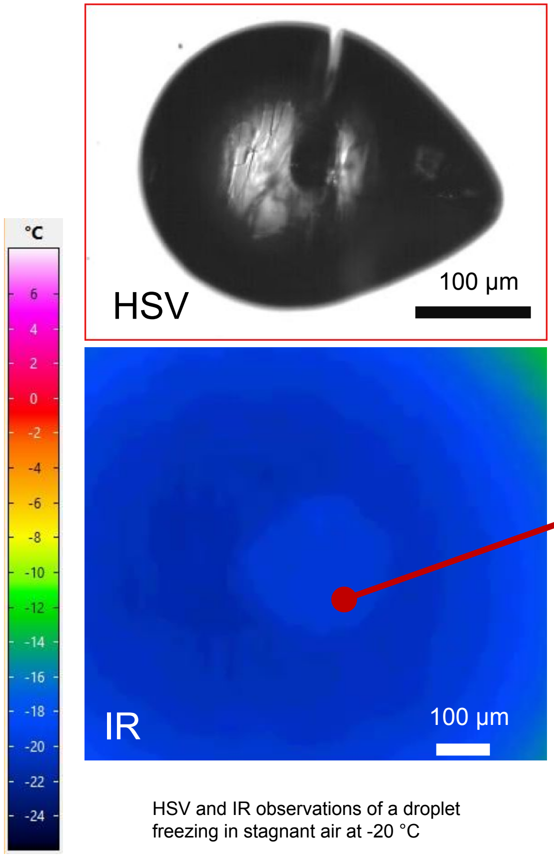


Stagnant air

(Lauber et al., 2018, JAS)

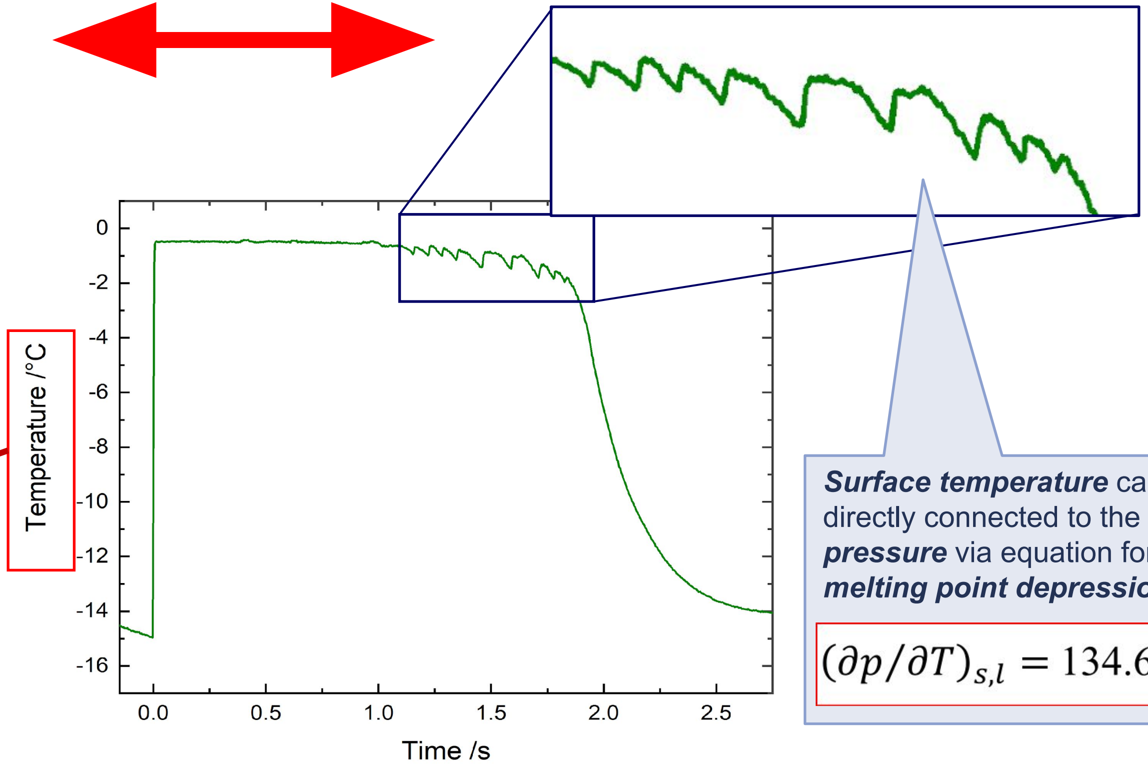


Synchronous IR and HSV imaging of freezing droplets



HSV and IR observations of a droplet freezing in stagnant air at -20 °C

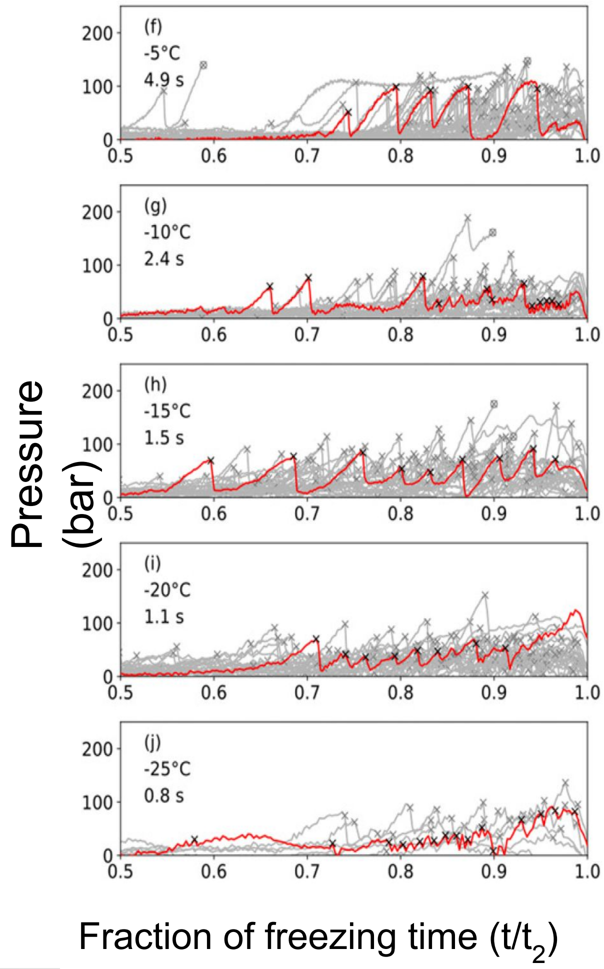
PRE = Pressure Release Events

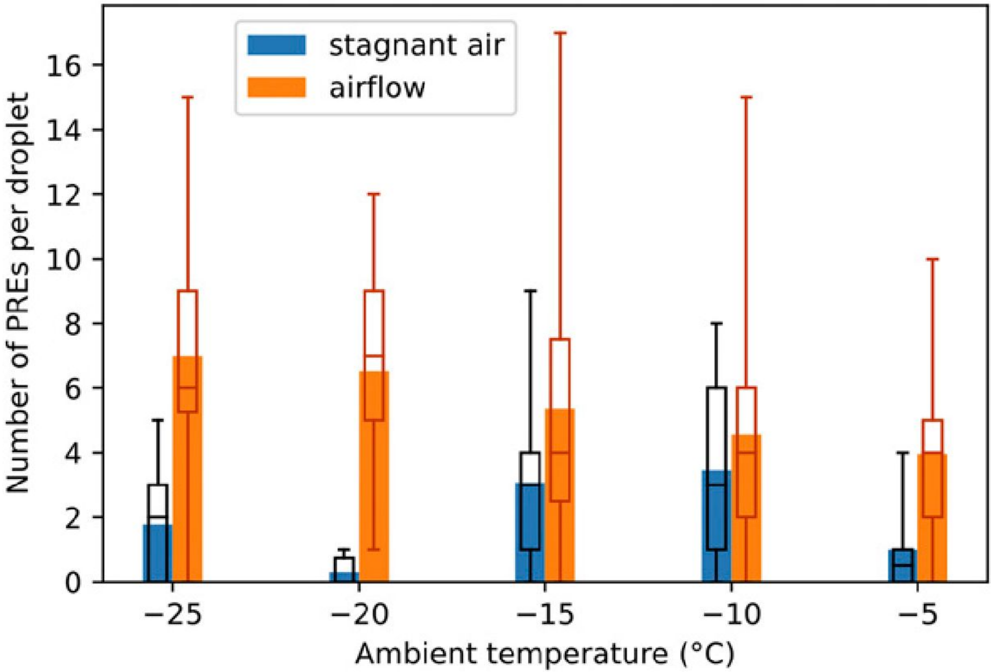


Surface temperature can be directly connected to the internal ***pressure*** via equation for the ***melting point depression***:

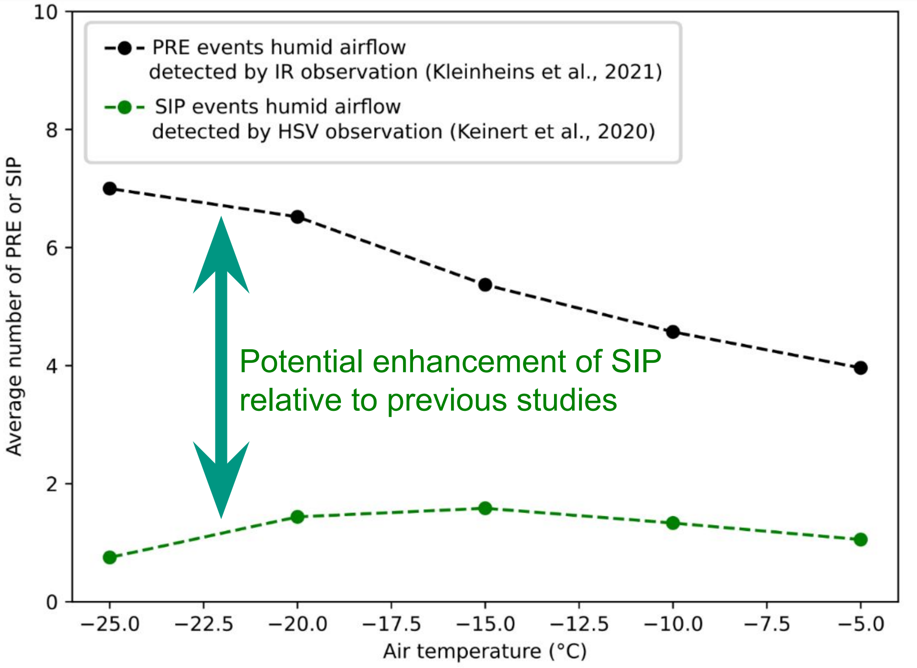
$$(\partial p/\partial T)_{s,l} = 134.6\ \mathrm{bar/K}$$

More frequent pressure release events (PRE) for droplets freezing in free fall





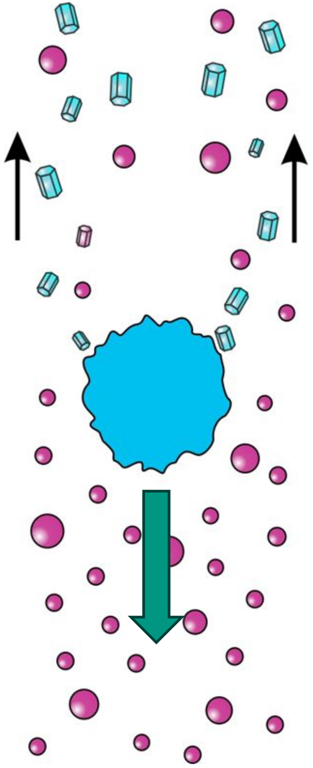
Open question (very urgent!):
Is every PRE really produce a secondary ice particle?
Detection of tiny subvisible ice particle urgently needed!

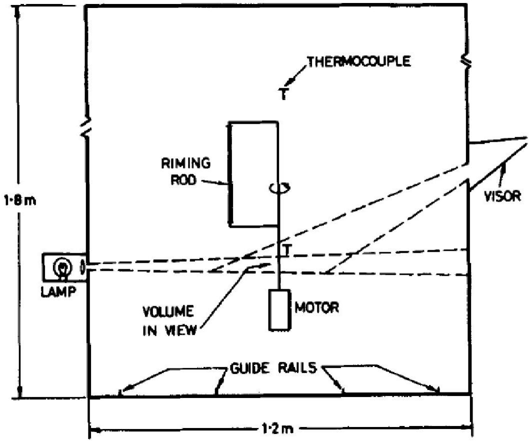


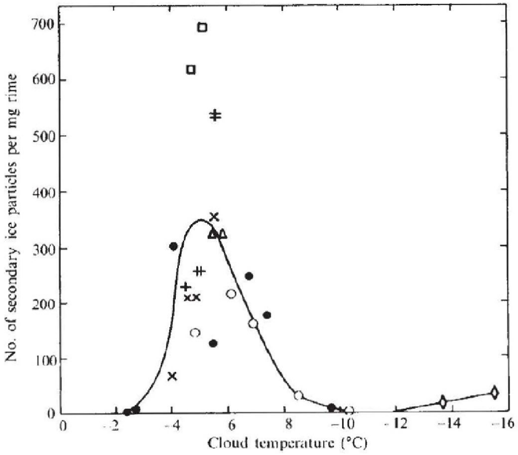
Lab studies example 2.
Rime-splintering SIP
(Hallett-Mossop mechanism)

Hallett and Mossop (Nature, 1974)
observed splinter formation during riming in a cloud chamber with LWC = 1 g/m^3 and droplet concentration of 500 cm^{-3}. They found that splinter production is active in the temperature range $-8°C < T_a < -3°C$ and it has a pronounced maximum at $T_a = -5°C$ and the drop impact velocity of 2.5m/s. With these conditions: 1 splinter per 250 droplets with D >24μm, or 700 splinters per mg rime

Physical mechanism under debate!!!







IDEFIX: Ice Droplets splintEring on FreezIng eXperiment

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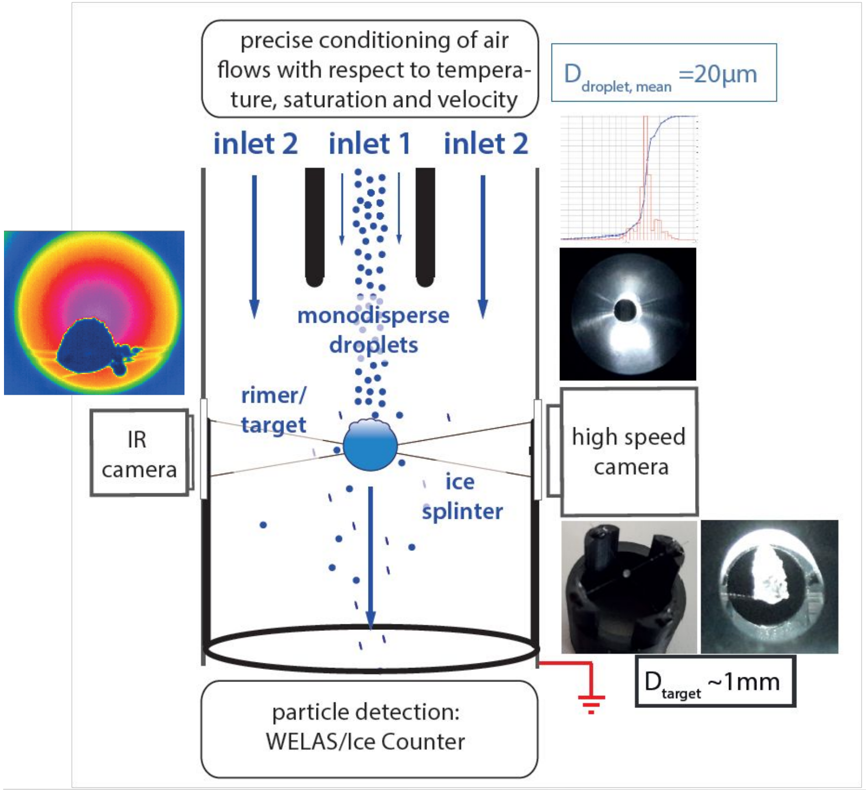
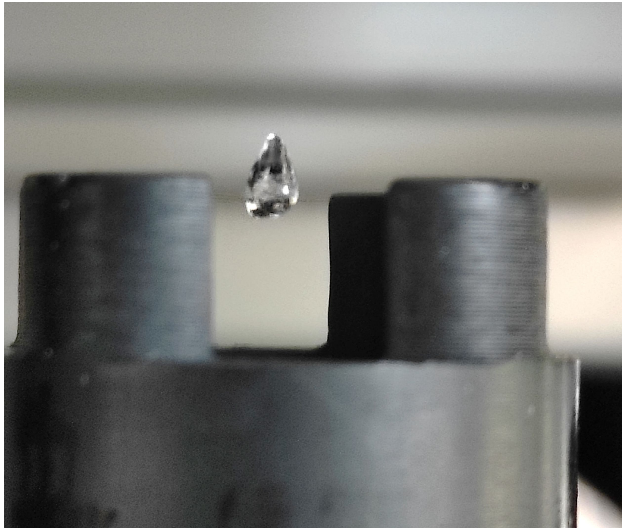
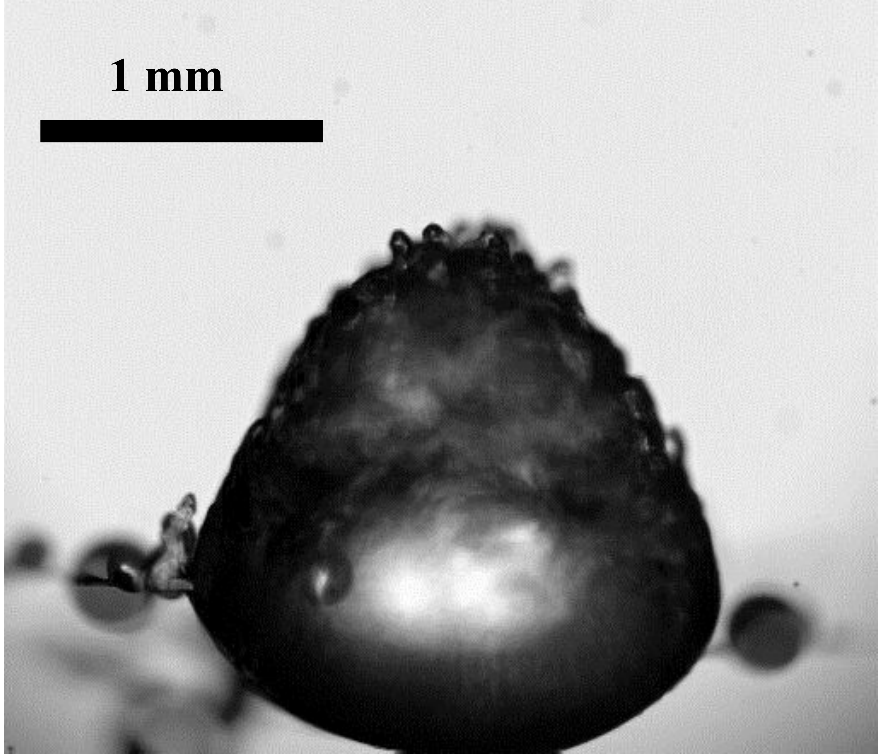


Experimental setup at TROPOS built by Susan Hartmann and Johanna Seidel (Seidel et al., ACP 2024).

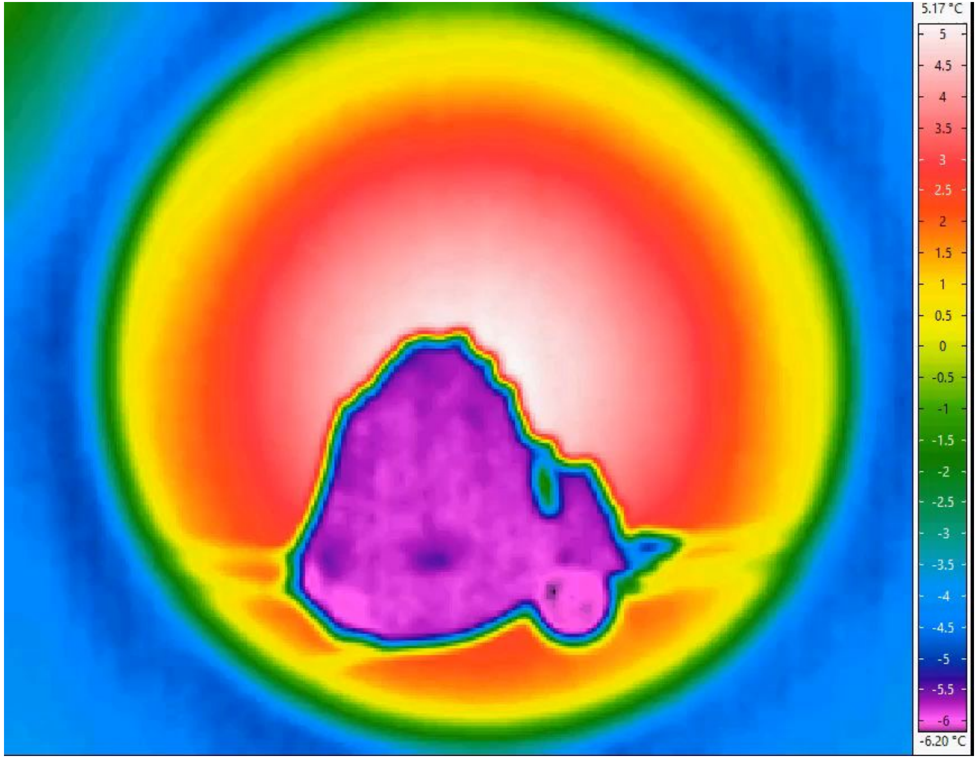


Rimed ice target at the end of 5 min riming

Riming ice target at -5°C, air flow velocity 1 m/s, mean drop. diameter 20 µm

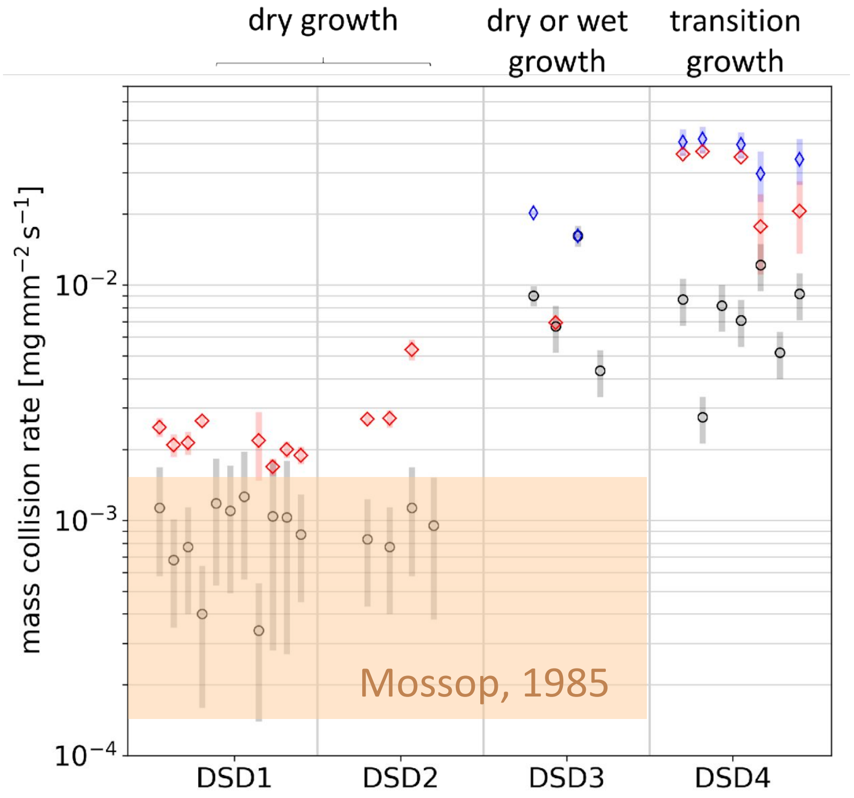


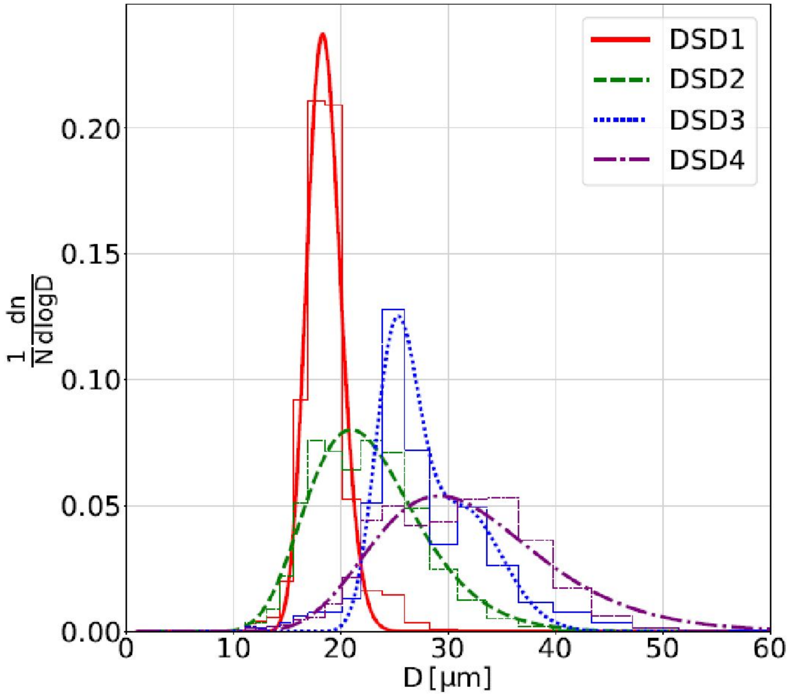
Target 20, appr. -6°C, after 5 min of riming

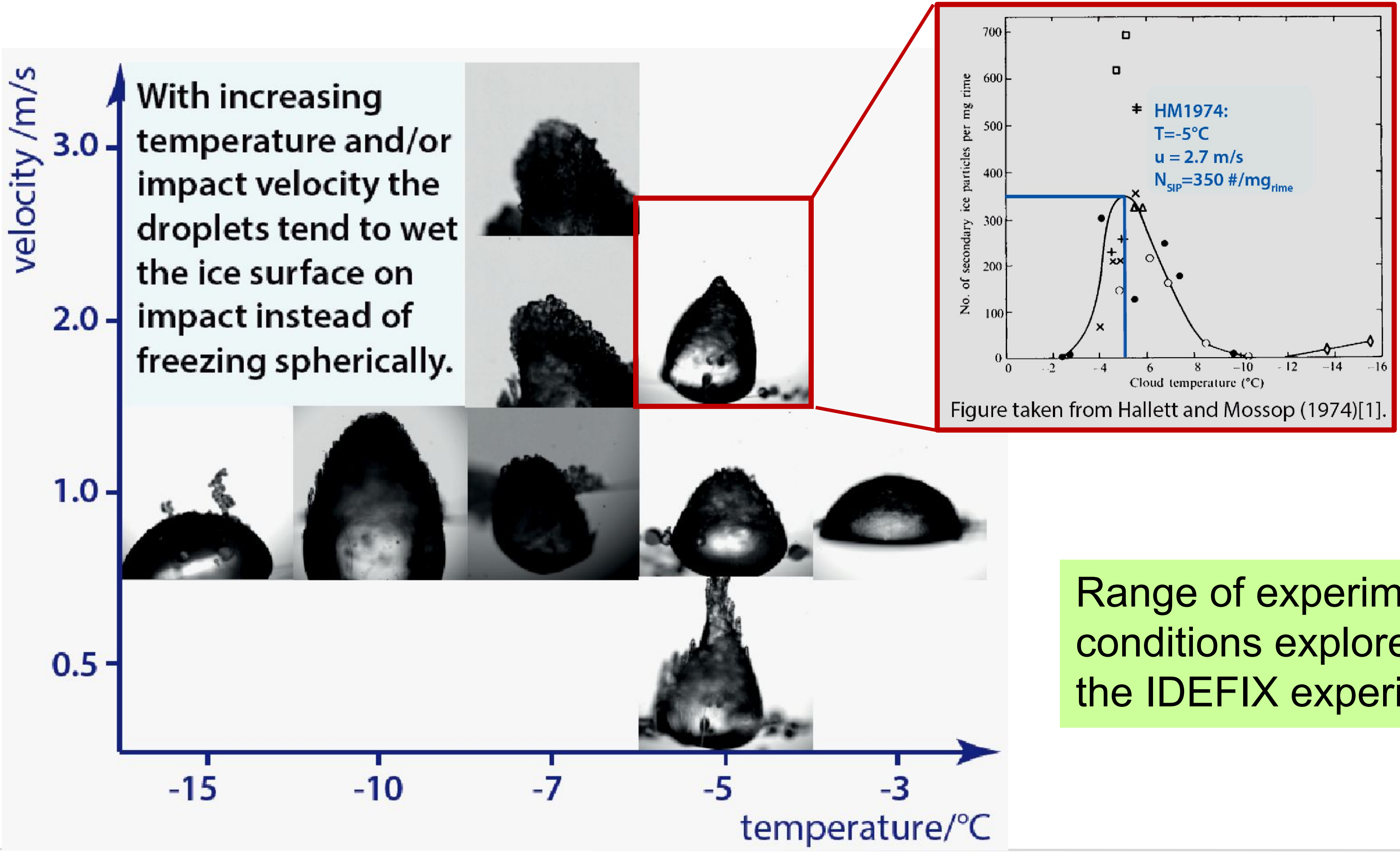


Target 20, IR record with 100 frames/s

The riming rate agrees well with realistic cloud conditions and Hallett-Mossop experiments







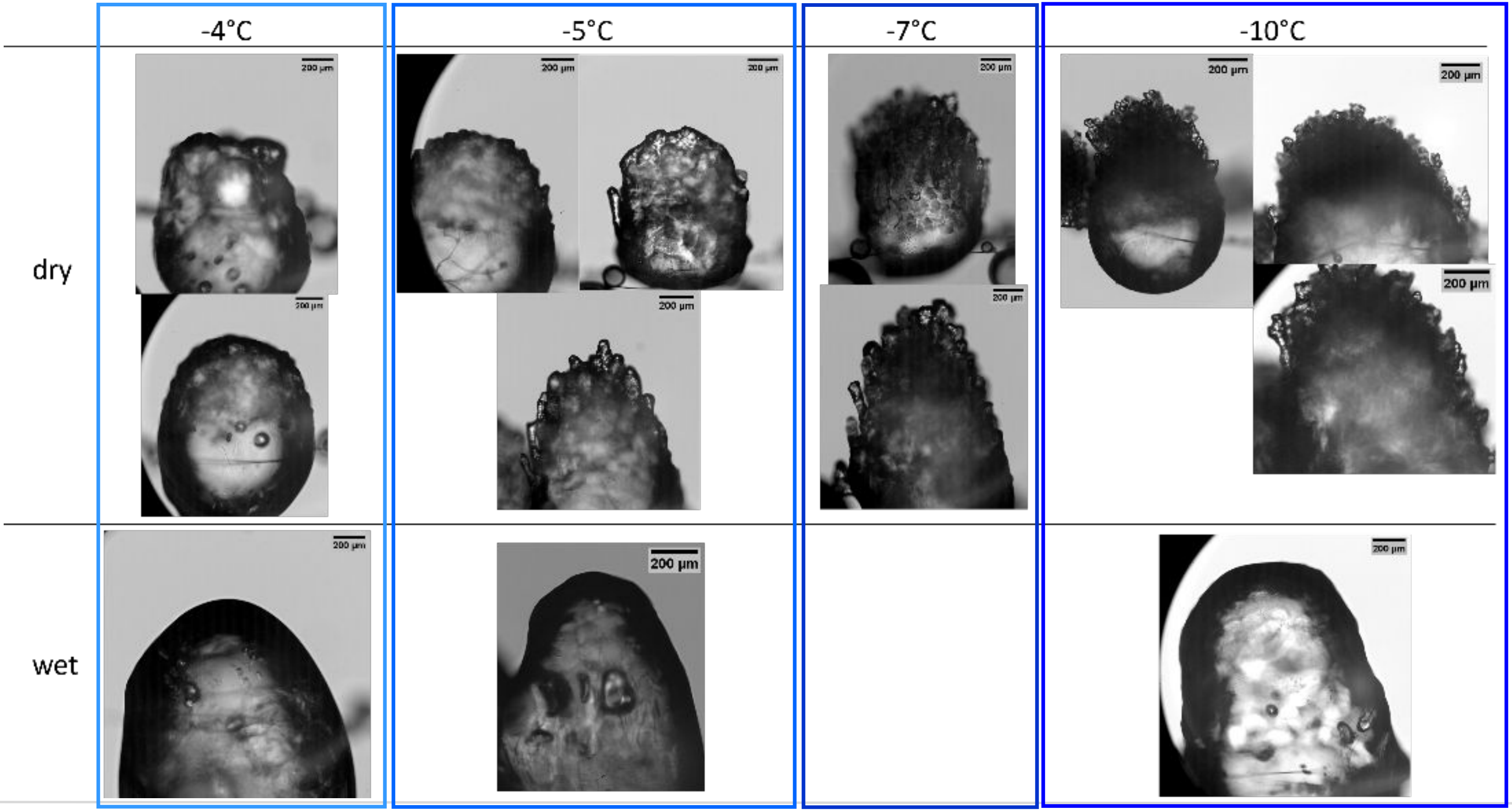
Range of experimental conditions explored in the IDEFIX experiment

No evidence of efficient

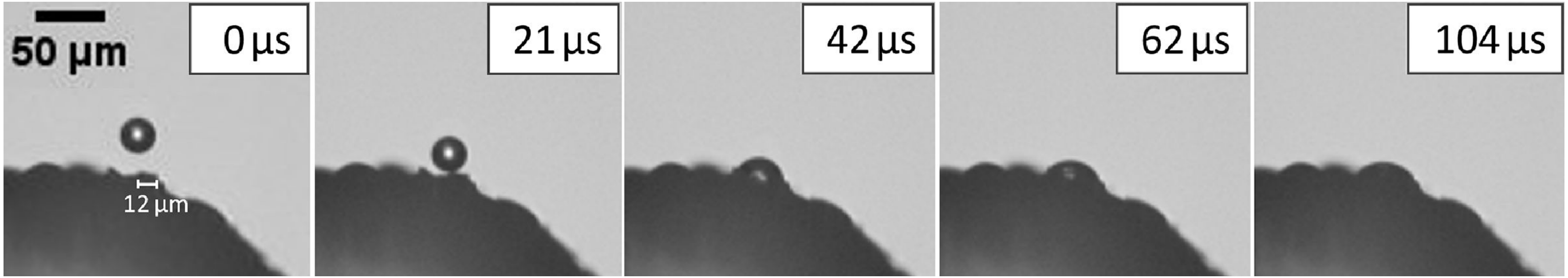
rime-splintering ice multiplication

in any of the experiments

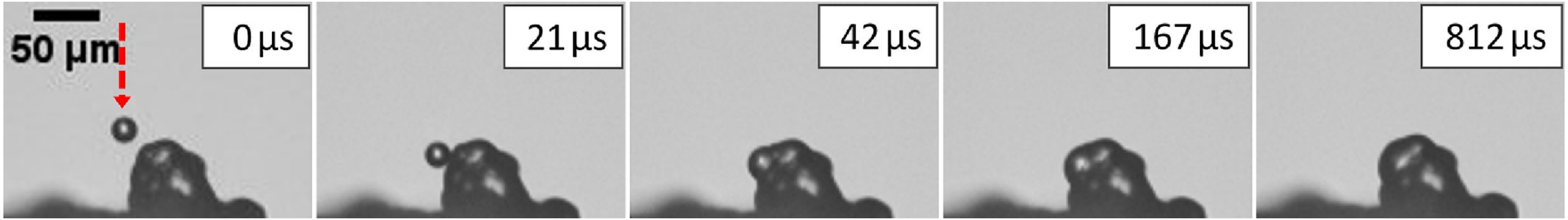
Realistic-looking rimed “graupel”



Potential SIP mechanisms: shell-fracture and glancing collision



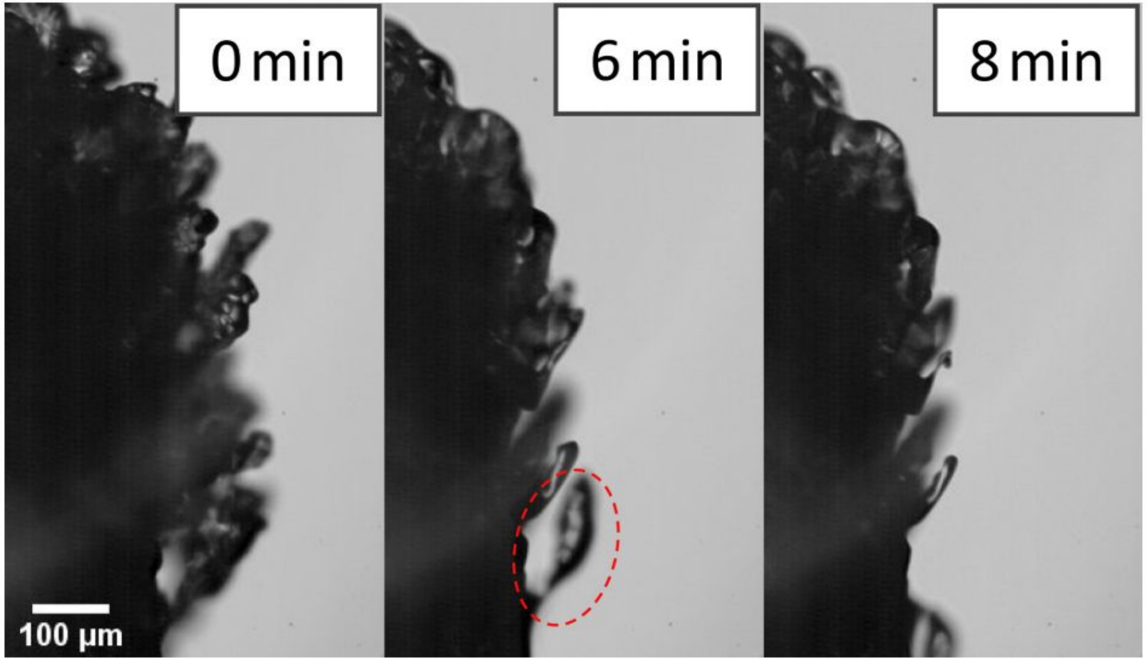
Shell-fracture hypothesis NOT CONFIRMED: 25 µm droplet freezing on a 12 µm wide ice neck at -7 °C and 1 m/s does not have a spherical shape



Glancing collision hypothesis NOT CONFIRMED: 20 µm droplet at -7 °C and 1 m/s is accreted
instead of bouncing off

Potential SIP mechanism: detachment of ice fragments due to sublimation

Sub-saturation conditions at relatively low temperature needed, the process is very slow.



Summary of recent lab studies

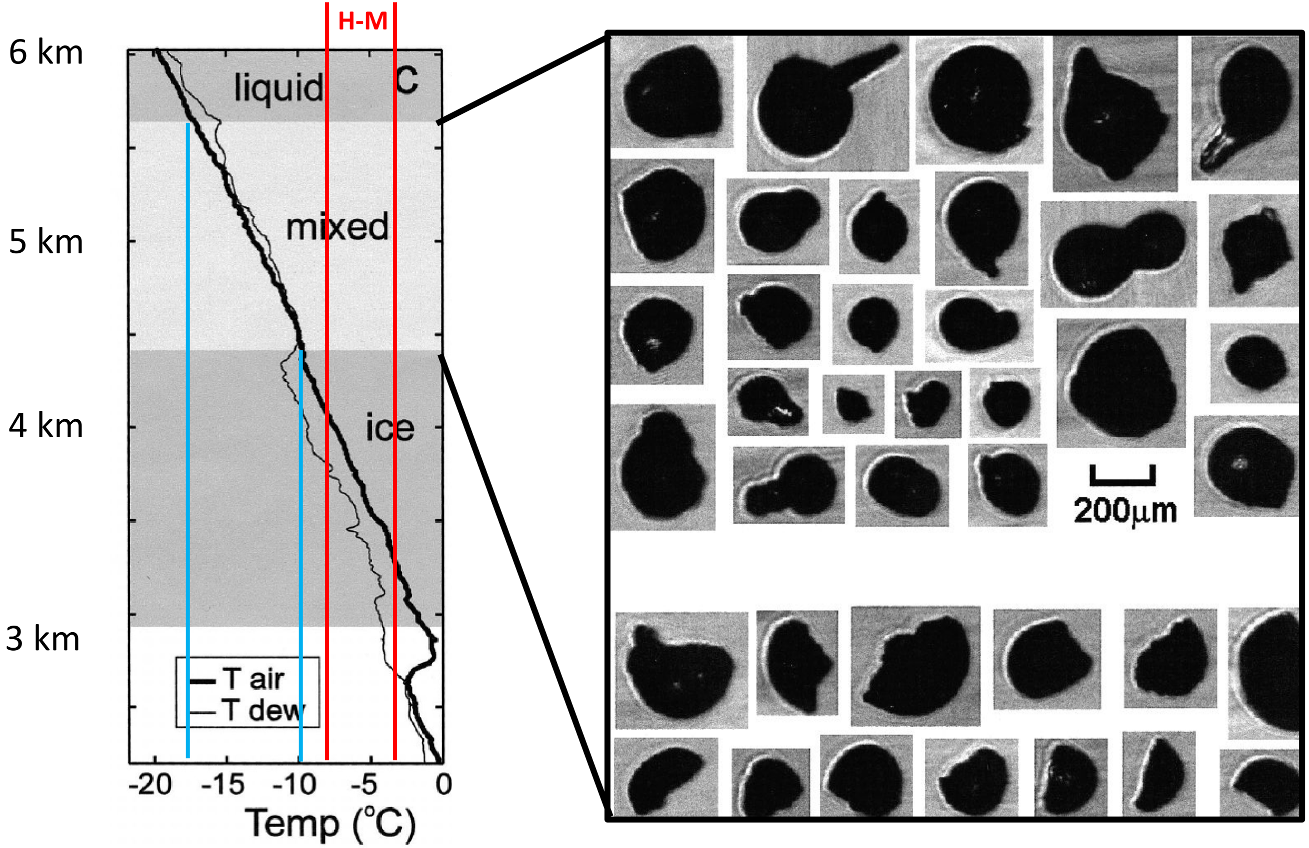
1. Lab experiments are essential for understanding the mechanisms of ice multiplication, but are **severely underrepresented** in the bulk of the SIP studies

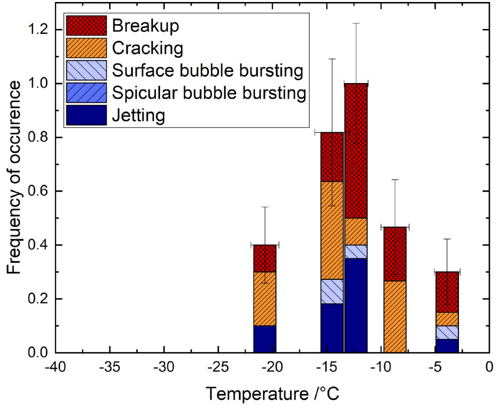
2. Current understanding of some mechanisms (e. g. droplet fragmentation upon freezing) is improving but some question are still open:
   a) **Number of small SI** particles produced in a PRE is unknown
   b) **Size dependence** must be quantitatively characterized

3. Efficient rime-splintering SIP could not be reproduced in the experimental setup:
   a) Neither **layer-fracture** nor **shell-fracture** hypothesis could be confirmed
   b) **Sublimation detachment of rime** not confirmed
   c) **Spherical freezing** on rough surface not confirmed
   d) **Freezing on a glancing contact** not confirmed

Do in-situ airborne observations support lab results?

Observation of the frozen drizzle droplets in a mixed phase cloud (AS-NS)

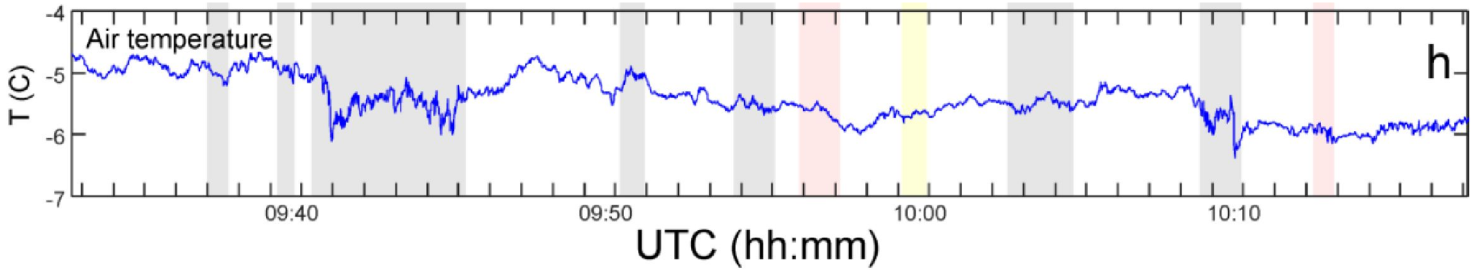




Korolev et al., JAM 2004:

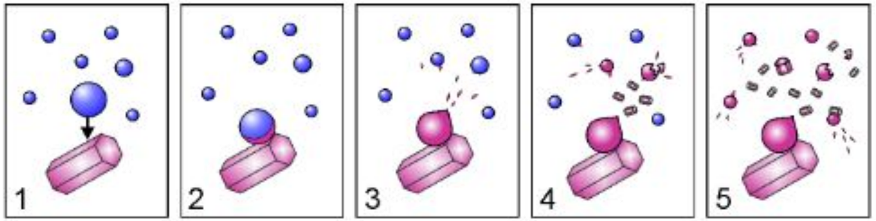
Shadow images of frozen droplet fragments collected during Canadian Freezing Drizzle Experiment, over Lake Ontario.

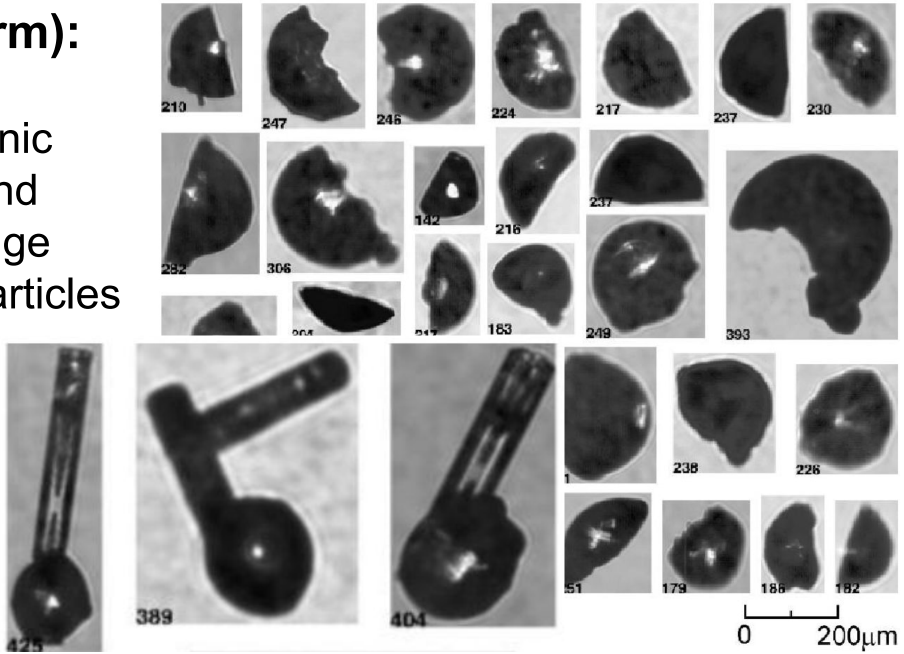
Instruments: CPI, Nevzorov probe.



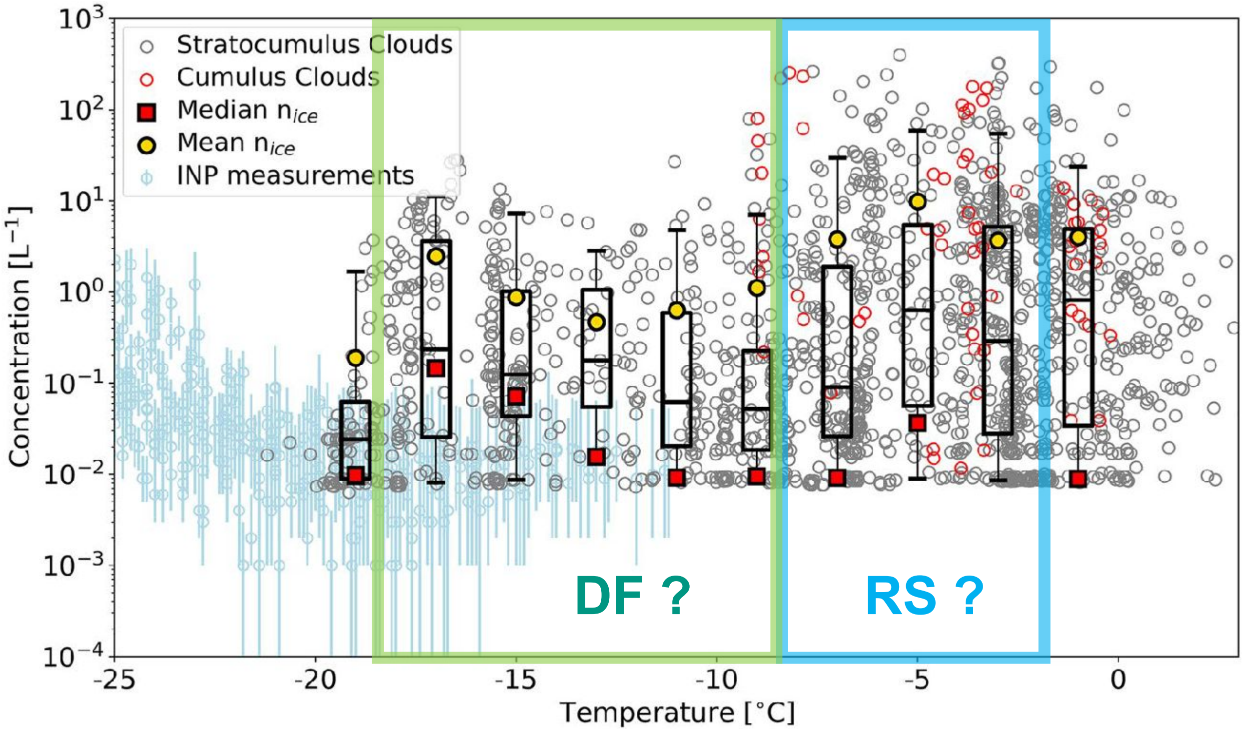
Same evidence, different conditions (warm):

Korolev et al., ACP 2019: Observations in oceanic tropical mesoscale convective systems (MCS) and mid-latitude frontal clouds in the temperature range from 0°C to -15°C heavily seeded by aged ice particles





Evidence for Secondary Ice Production in Southern Ocean Maritime Boundary Layer Clouds (SOCRATES, Järvinen et al., JGRA 2022)



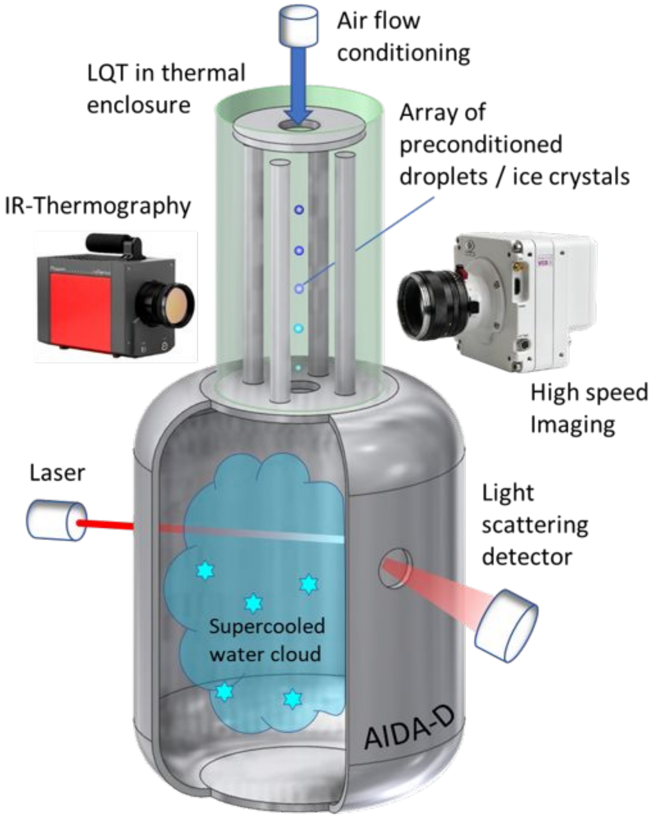
Blue circles: measurements of INPs from the HIAPER taken both below and above the sampled clouds

How to move forward?

1. Ice multiplication mechanisms cannot be fully simulated **in the lab** because the conditions are not nearly realistic.

   Solution: improve control of process parameters in experiment, focus on the mechanistic understanding instead of trying to reproduce the whole process (which cannot be done anyway). That could eventually help excluding non-physical interpretations thus leading to a better design of future experiments / field campaigns.

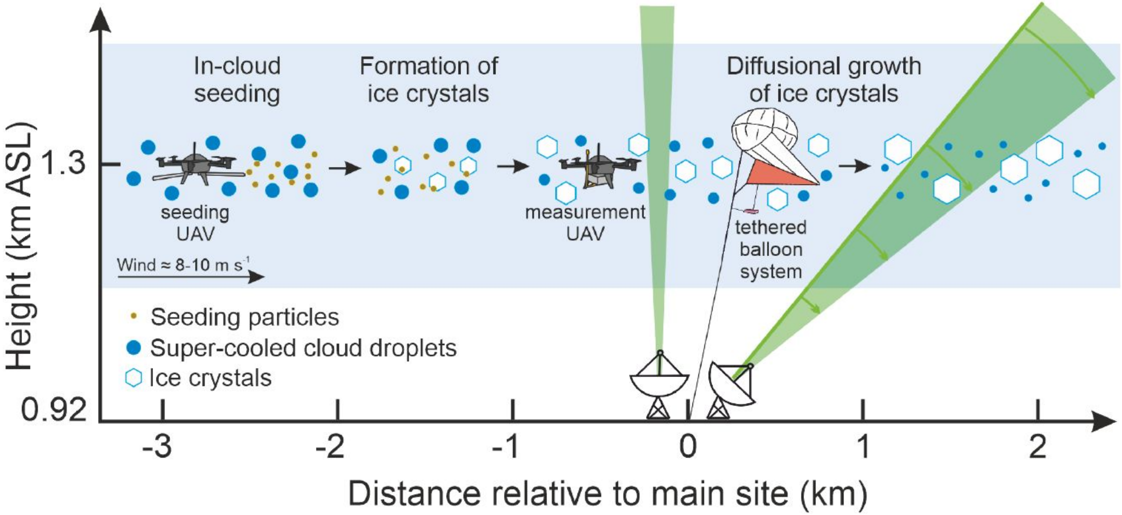
   Experimental studies involving cloud chambers?

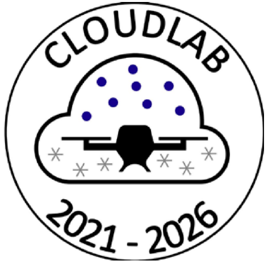


How to move forward?

2. Ice multiplication cannot be unraveled by **in-situ** methods (aircraft-based observations) alone because the SIP events are too fast and too rare. Indirect methods required.

Solution: 1) combine aircraft and remote observations;
2) induce SIP events artificially! A real cloud is the best cloud lab.





Conducted **70 glaciogenic seeding experiments** of stratus clouds from a UAV
(Henneberger and Ramelli et al., BAMS, 2023; Miller et al., ACP 2024; Miller et al., CRST, 2024)

Thank you for your attention!

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…and this one too.