Experimental and modeling study of heterogeneous ice nucleation on mineral aerosol particles and its impact on a convective cloud

thesis conducted under the supervision of
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Thibault Hiron[1,2] | September 29, 2017

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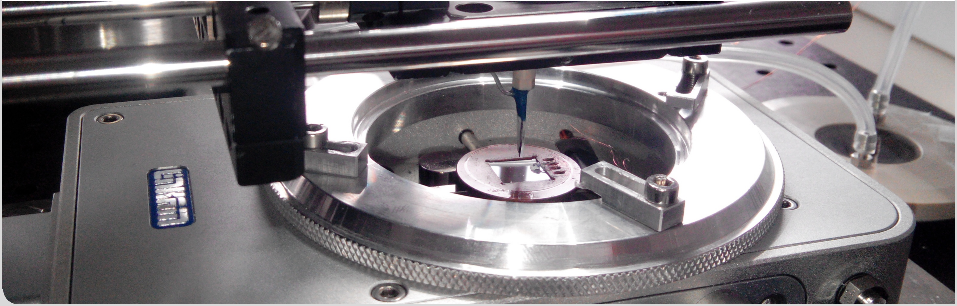
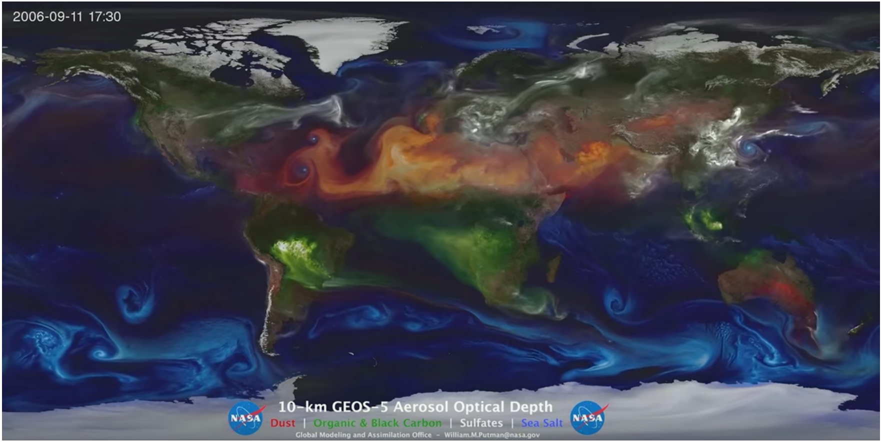


Introduction

APs: small particles ($10\,\mathrm{nm}$–$10\,\mathrm{\mu m}$) in suspension in the atmosphere.



Global repartition of aerosol particles at $10\,\mathrm{km}$ of altitude.

Different natures: Soluble/insoluble, hydrophillic/hydrophobic, smooth/rough, ...

Introduction

Formation of clouds

- Clouds contain droplets of liquid water and ice crystals (hydrometeors)
- Water vapor needs aerosol particles (AP) to form hydrometeors

Aerosol role in clouds formation

Two means of influence for aerosol particles

- Base for water vapor condensation into droplets
  $\Rightarrow$ Cloud Condensation Nuclei (CCN)
- Base for development of ice crystals
  $\Rightarrow$ Ice Nuclei (IN)

CCN role quite well understood but IN role under intense research:
Major impact of ice nucleation on clouds lifetime and precipitations

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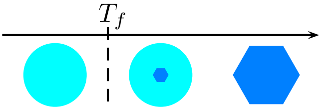
At low temperatures, the water in the droplets change phase: it is the ice nucleation.



Ice nucleation ⇒ Phase change:

- Reorganization of the molecules into a crystal (critical germ formation and creation of an interface);
- Growth of the crystal.

This yields an energy barrier ⇒ metastable state of water below $0°\mathrm{C}$.
Droplets of pure water freeze at temperatures generally below -36$°\mathrm{C}$
⇒ **Homogeneous nucleation**.

In the atmosphere, ice nucleation observed at temperatures up to $-5°\mathrm{C}$
⇒ homogeneous nucleation cannot explain these high temperatures!

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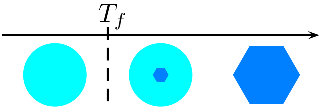

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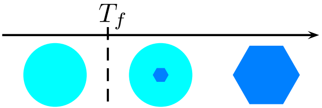

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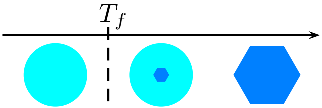

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Ice crystals observed at "*high*" temperatures
$\Rightarrow$ lowering of the energy barrier.
Insoluble aerosol particles can reduce the energy of critical radius formation.
$\Rightarrow$ **Heterogeneous Nucleation**

Ice Nuclei

Aerosol particles capable of lowering the energy of germ formation.
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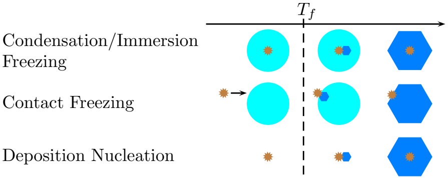

Heterogeneous ice nucleation mechanisms

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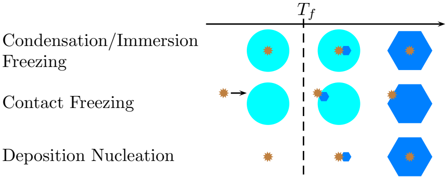

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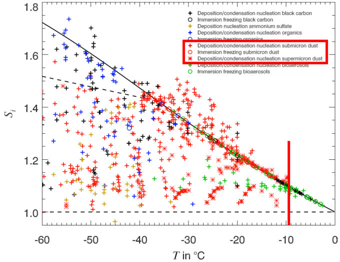
Mineral particles

- Distributed all over the globe;
- Known to be rather good IN;
- Onset data points spread across the phase diagram.

Recently: Feldspar identified as highly efficient ice nucleus in immersion freezing mode (*e.g.* Atkinson, 2013; Harrison, 2016; Peckhaus, 2016).



Overview of the nucleation onset temperatures and saturation ratios.
Source: Hoose and Möhler, 2012

Single point of view in experiments

Feldspar intensively investigated in the immersion freezing mode but very few deposition nucleation experiments (Yakobi–Hancock, 2013; Kiselev, 2017).

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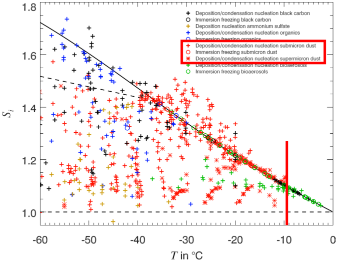

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Quantify the role of mineral particles on ice and precipitation formation

Two different methods with specific questions: Experiment & Modeling.

Experimental study: Cold Stage, IMK–AAF

For mineral particles, does being a good immersion freezing ice nucleus imply being a good deposition nucleation ice nucleus?

Modeling study: Descam, LaMP

What is the impact of the different ice nucleation mechanisms, involving mineral aerosol particles, on the development of a convective cloud?

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Outlook

1. Cold Stage
   - Motivation
   - Experimental Setup
   - Experiments with K-Feldspar
   - Analysis
   - Summary
2. Case Study of ice nucleation on minerals using DESCAM
   - Motivation
   - Preliminary study
   - Model development
   - Results
   - Discussion
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Cold Stage

Scientific objectives

- Study immersion freezing and deposition nucleation on mineral particles:
  ⇒ K-Feldspar identified as very active and widely investigated;
  (Atkinson, 2013; Yakobi–Hancock, 2016; Peckhaus, 2016; Kiselev, 2017)
- Look into links between immersion freezing and deposition nucleation;
- Develop parametrizations for ice nucleation;
- Ideally, aim for a joint parametrization for immersion freezing and deposition nucleation.

Requirement

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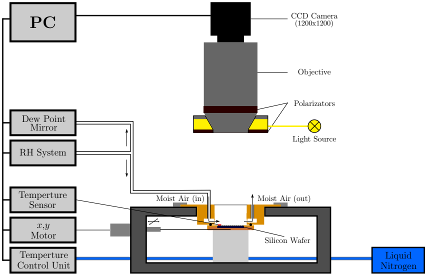
Base unit

- Linkam motorized Cold Stage
- Droplets of particles suspensions deposited on a silicon wafer
- Simultaneous observation of 150 droplets



Development of a flow cell fixed on top of the cold stage

Reduced volume and improved homogeneity of the flow field

Humidity control system

Dew point in $[-70°\mathrm{C}$ to $4°\mathrm{C}]$, air flow of $150\,\mathrm{mL.min}^{-1}$

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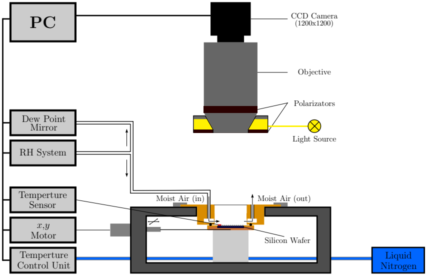

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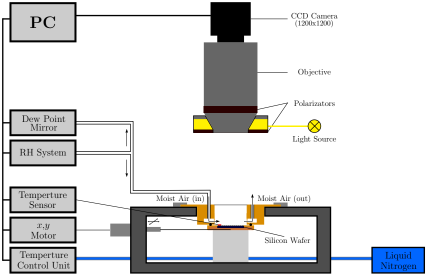

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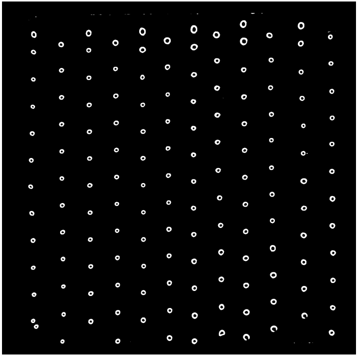
Experiments with K-Feldspar

Principle of experiment

- Consecutive nucleation cycles in immersion freezing and deposition nucleation mode;
- Suspension of K-Feldspar particles deposited in an array of *ca.* 150 droplets of 0.6 nL
- Three different concentrations for broad temperature range.



Wafer at first frame of experiment

Suspension	A	B	C
Concentration $(\mathrm{g/L})$	$(2.5 \pm 0.1) \cdot 10^{-1}$ C_0	$(2.5 \pm 0.2) \cdot 10^{-2}$ $C_0/10$	$(2.5 \pm 0.2) \cdot 10^{-3}$ $C_0/100$

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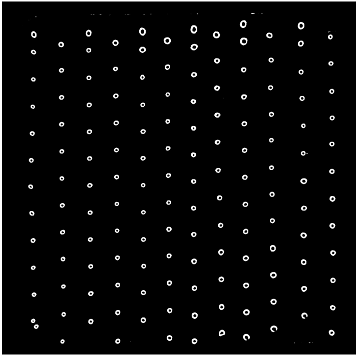

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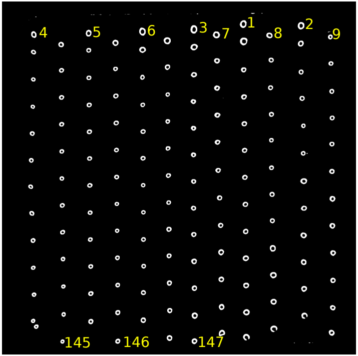

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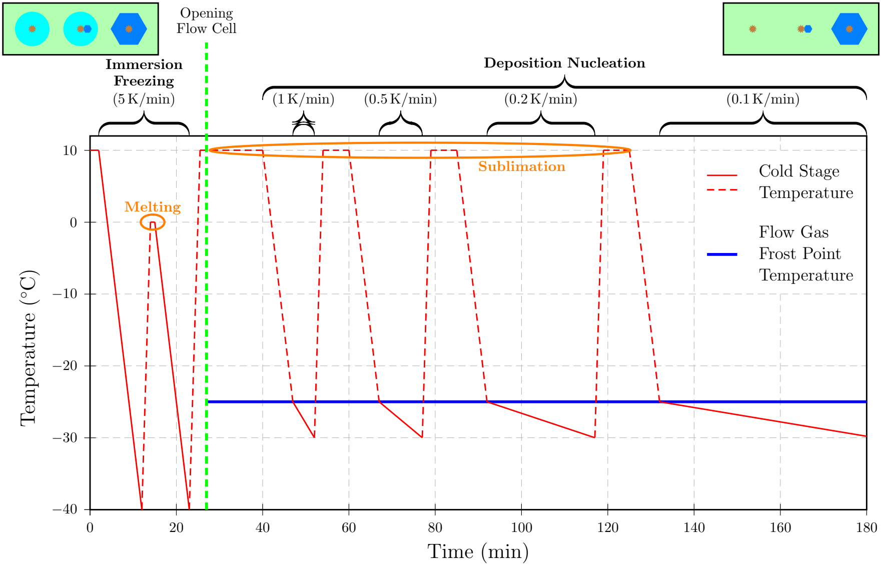

Experiments with K-Feldspar

For each different suspension, freeze-thaw cycles at a cooling rate of $5\,\mathrm{K/min}$.

- Suspension **A** $(2.5 \cdot 10^{-1}\mathrm{g/L})$: 2 cycles;
- Suspension **B** $(2.5 \cdot 10^{-2}\mathrm{g/L})$: 3 cycles;
- Suspension **C** $(2.5 \cdot 10^{-3}\mathrm{g/L})$: 3 cycles.

Freezing temperature decreases with decreasing total surface of aerosol particle per droplet.
For suspensions **A** and **B** similar curves for the different cycles.

For suspension **C**, apparent enhancement of freezing efficiency below $-30°\mathrm{C}$ but similar freezing curves above this temperature.
$\Rightarrow$ First 15 frozen droplets show similar behavior in each cycle.

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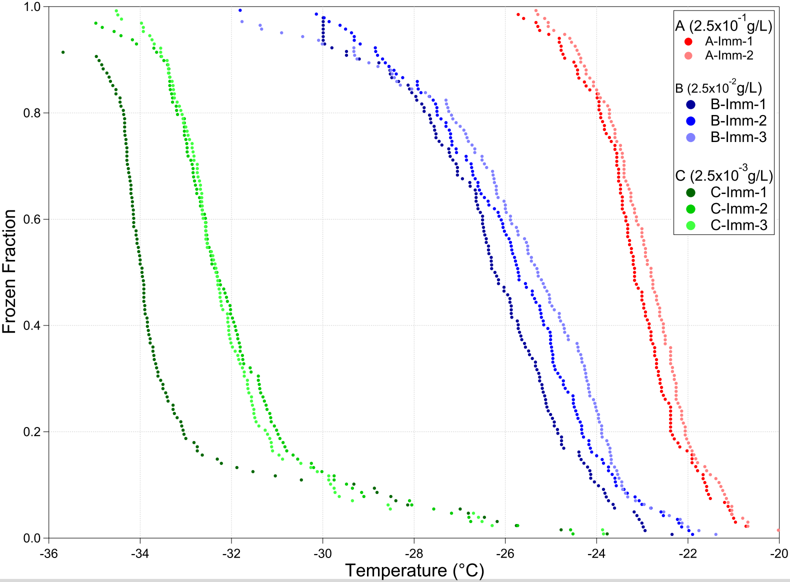

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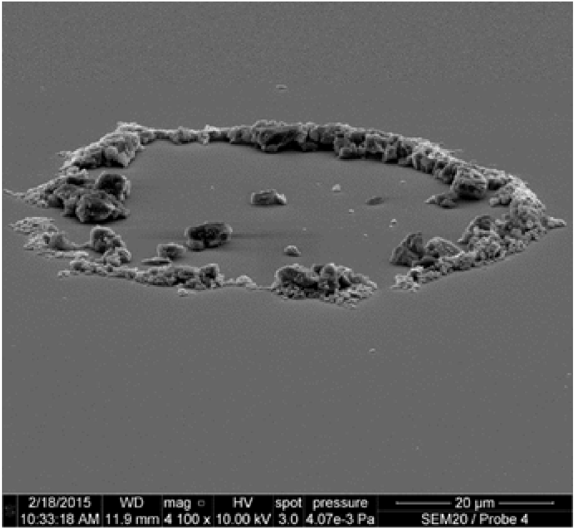
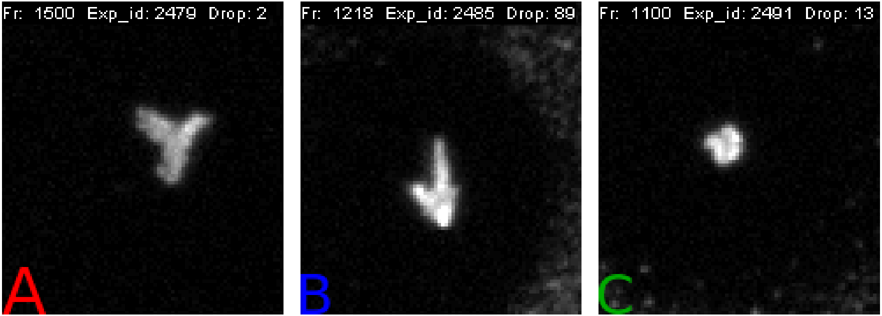


Residuals from a droplet on a silicon wafer, courtesy of A. Kiselev (IMK/AAF)

Experiments with K-Feldspar

Number of ice nucleating sites

- **A** and **B**: several ice nucleation sites per residuals (left and center);
- **C**: only one ice nucleation site per residuals (right).



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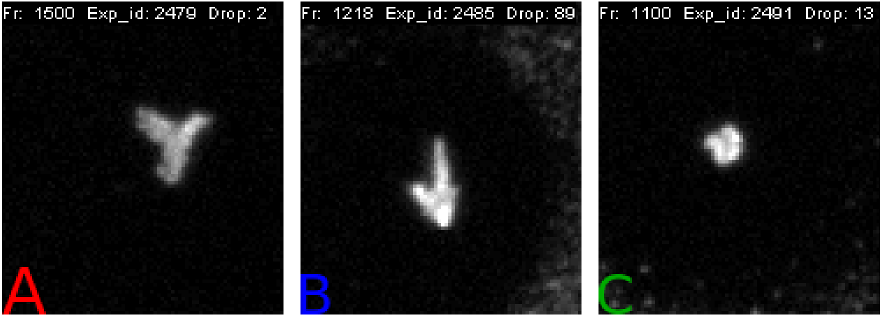

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Droplets involved in ice nucleation

Comparison of the 15 first droplets frozen via immersion mechanism with the 15 first residuals that nucleated ice in deposition mode ($(1\mathrm{K}/\mathrm{min})$):

- C-Imm-1: [46, 15, 13, 37, 77,103,138,136,124, 58, 52, 82, 96, 33, 89]
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Same ice nucleating sites for different experiments but in different orders.

Link between IN activity in immersion freezing and deposition nucleation modes or preactivation of active sites?

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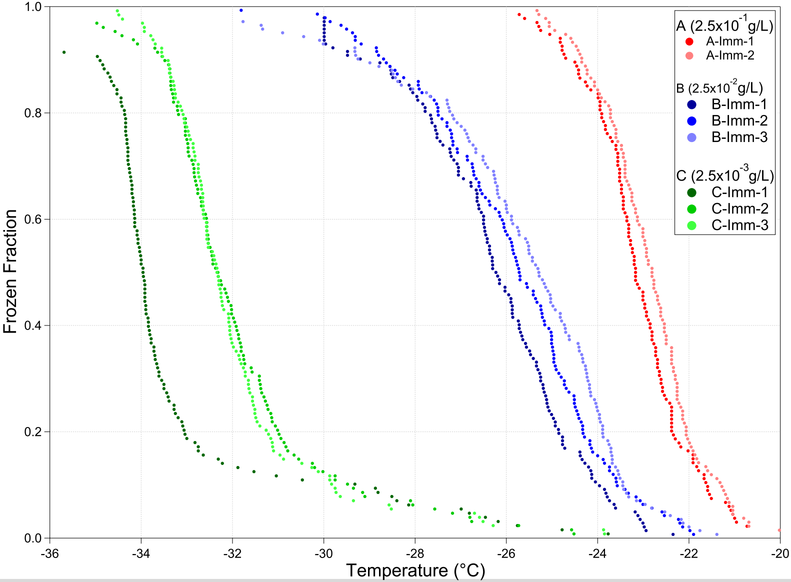

Frozen Fraction
1.0
0.8
0.6
0.4
0.2
0.0
-36
-34
-32
-30
-28
-26
-24
-22
-20
Temperature (°C)
A (2.5x10^{-1}g/L)
A-Imm-1
A-Imm-2
B (2.5x10^{-2}g/L)
B-Imm-1
B-Imm-2
B-Imm-3
C (2.5x10^{-3}g/L)
C-Imm-1
C-Imm-2
C-Imm-3

Analysis

Droplets involved in ice nucleation

Comparison of the 15 first droplets frozen via immersion mechanism with the 15 first residuals that nucleated ice in deposition mode ($1\mathrm{K}/\mathrm{min}$):

- C-Imm-1: [46, 15, 13, 37, 77,103,138,136,124, 58, 52, 82, 96, 33, 89]
- C-Imm-2: [15, 46, 13,138, 77, 76, 37,103, 58, 96, 16,132, 33, 26,136]
- C-Imm-3: [46, 15, 13, 77,136, 37, 58,138, 96, 74, 82, 39, 99, 88,103]
- C-Dep-2: [15, 13, 46, 37, 42, 58, 96,103,136,133, 77, 33, 82,104,132]
- C-Dep-3: [116, 42, 46, 60, 77, 15, 53, 13,104,103, 87, 96, 78, 37,138]
- C-Dep-4: [77, 13, 49, 46,116, 96,103, 15, 18, 95, 32, 60, 58, 90, 87]

Same ice nucleating sites for different experiments but in different orders.

$\Rightarrow$ Link between IN activity in immersion freezing and deposition nucleation modes ~~or preactivation of active sites~~.

Cold Stage

Scientific summary

- Heterogeneous nucleation takes place on specific sites in both nucleation modes;
- Resulting INAS density parametrization for K-Feldspar particles:
  - $n_{s,\mathrm{dep}}(T, RH_i) = \exp(11.2 - 0.18 \cdot T) \cdot [1 - \exp(0.02 \cdot (127 - RH_i))]$
  - $n_{s,\mathrm{imm}}(T) = \exp(10.3 \cdot \exp[-\exp(0.345 \cdot (T + 21.2))] + 6.05)$
- Consecutive immersion freezing and deposition nucleation experiment *on the same identified* particles:
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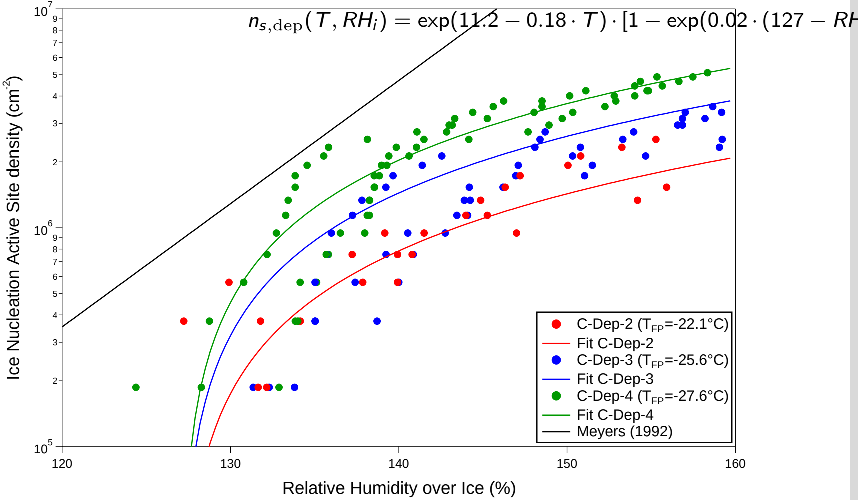

Ice Nucleating Active Site density as a function of temperature in deposition nucleation mode

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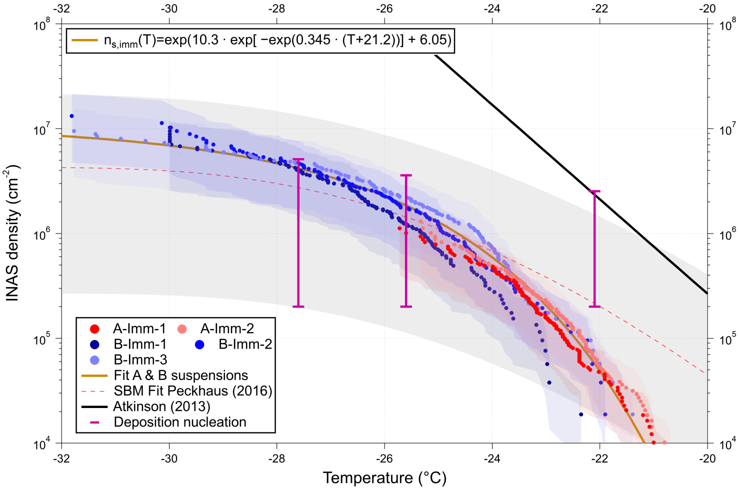

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Outlook

1. Cold Stage
   - Motivation
   - Experimental Setup
   - Experiments with K-Feldspar
   - Analysis
   - Summary
2. Case Study of ice nucleation on minerals using DESCAM
   - Motivation
   - Preliminary study
   - Model development
   - Results
   - Discussion
   - Summary

Descam

Scientific objective

- Study the role of the different ice nucleation mechanisms on the development of a convective cloud;
- Study the impact of different types of mineral aerosol particles.

Technical requirement

"[improve] *the treatment of the microphysical ice processes*" (Leroy, 2006)

Modeling studies

- Preliminary modeling study for methodology, using standard parametrizations;
- Case study with improved treatment of ice nucleation;
- Implementation of Cold Stage experiments parametrizations.

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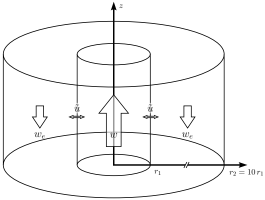

Dynamic Framework

- 1D5 cylindrical model (Asai and Kasahara, 1967)
- Case study: CCOPE (Dye et al, 1986, Leroy et al, 2006)

A bin resolved model

We follow the explicit spectra for:

- aerosol particles, cloud droplets and ice crystals numbers;
- mass of aerosol in each category.

In total 6 size distributions.

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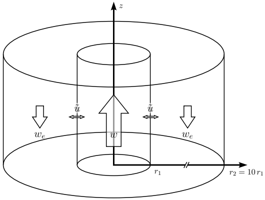

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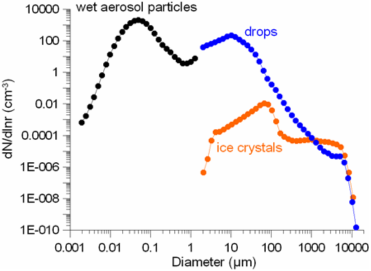

Size distributions in DESCAM

Descam: Preliminary study

Methodology (Hiron & Flossmann, JAS, 2015)

Theoretical studies of the ice formation mechanisms:

- considered separately (one mechanism considered per simulation) and in competition;
- pristine ice formed for each nucleation mechanism;
- respective impact on the dynamic of the cloud.

Parametrizations of ice nucleation

Homogeneous nucleation: Koop et al., 2000.

Heterogeneous nucleation:

- Immersion freezing: Bigg, 1956;
- Condensation freezing and deposition nucleation: Meyers, 1992;
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- High altitude steady cloud formation seems to be dominated by deposition nucleation;
- Strong influence of temperature regime and mode of ice nucleation on the cloud development.

The method allows the identification of governing parameters of the impact of ice nucleation on the cloud development.

Limitations

Simplified representation of ice nucleation:

- no dependence on the nature of the aerosol particles;
- no dependence on the size of the aerosol particles.

Heterogeneous nucleation parametrized with *old* parametrizations (1956 and 1992) $\Rightarrow$ new representations with more precision (INAS, CNT, ...)

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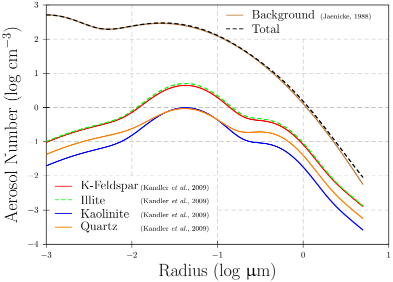
Descam: Model development

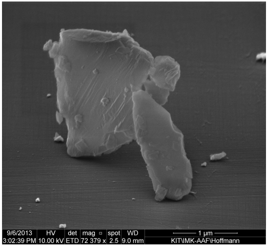
Saharan Desert Dust

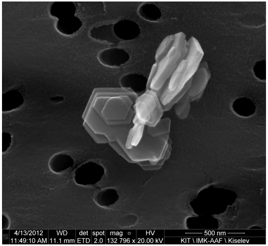
Four types of atmospherically relevant minerals:

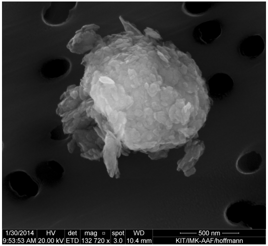
- (K-)Feldspar
- Kaolinite
- Illite
- Quartz

size distributions from Kandler, 2009 (*contra*)









SEM pictures of K-Feldspar, Kaolinite and Illite (from left to right) courtesy of N. Hoffmann & A. Kiselev (IMK-AAF/KIT)

Descam: Model development

Use of aerosol-specific ice nucleation parametrization:

- Ice Nucleating Active Site (INAS) densities, defined using the total aerosol particle surface per particle or per droplet (A_{aer}).

$\Rightarrow$ Supplementary information on aerosol particles in the model needed:

- Total surface of aerosol particles in each bin of the aerosol particles, droplets and ice crystals size distributions;
- Total number of aerosol particles in each bin of the droplets and ice crystals size distributions.

5 new size distributions, 4 new types of APs $\Rightarrow$ 47 distributions

- Number of aerosol particles [5]: $\mathcal{N}_a$, $\mathcal{N}_d$ and $\mathcal{N}_i$;
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Descam: Results

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INAS density for K-Feldspar from Atkinson *et al.*, 2013.

Results

Cloud starts to form around $3\,\mathrm{km}$ after $8\,\mathrm{min}$ and develops over $20\,\mathrm{min}$.

In–cloud ice becomes noticeable at $7\,\mathrm{km}$, entire cloud becomes frozen.

Precipitation starts after $34\,\mathrm{min}$ between 6 and $6.5\,\mathrm{km}$.

Latent heat release due to freezing delays precipitation onset.

$\Rightarrow$Large impact of immersion freezing on rain formation.

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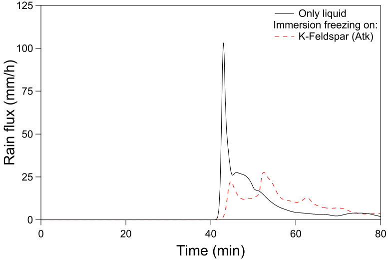

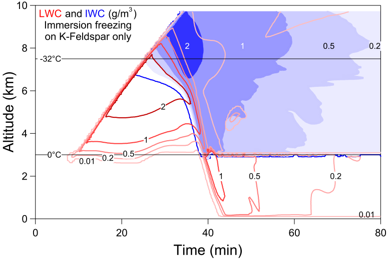

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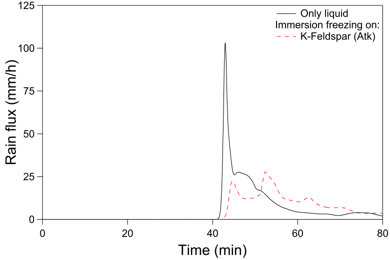

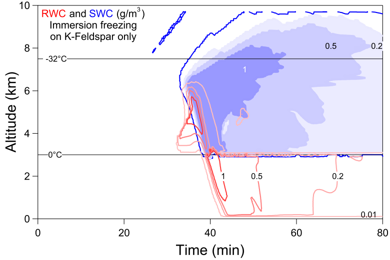

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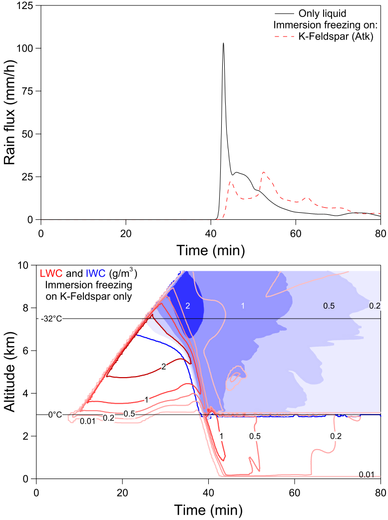

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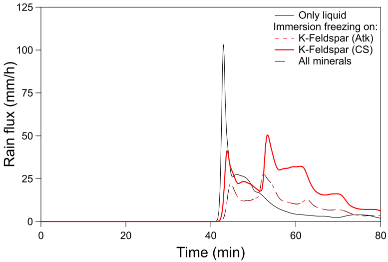

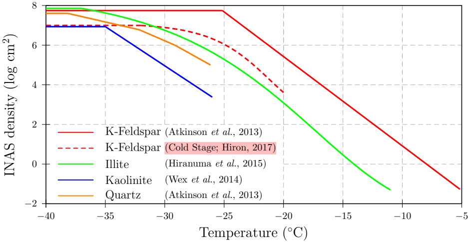

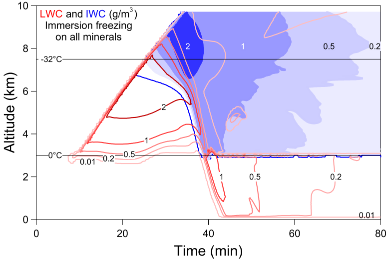

Results

Minerals active at warmer temperatures have a larger impact on the rainfall dynamics

K-Feldspar has the most impact among mineral IN

$\Rightarrow$ rainfall rate identical to case with all minerals.

Descam: Results

Heterogeneous nucleation parametrizations

Contact freezing: Hoffmann, 2015

Deposition nucleation:

- K-Feldspar: Cold Stage experiments; Hiron, 2017;
- Illite: Wheeler and Bertram, 2012;
- Kaolinite: Wex *et al.*, 2014

Results

- Almost no impact of deposition nucleation on the cloud dynamics, as most aerosol particles are activated into droplets;
- No impact of contact freezing on the cloud development, but large impact on in–cloud precipitating IWC $\Rightarrow$ increase in precipitation;
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Limitations

41 new size distributions
⇒ computing time expensive model
⇒ not applicable to more complicated dynamical frames, *eg.* Descam 3D.
Simplifications are needed!

Two algorithms for immersion freezing:

- Condensation freezing ⇒ only one aerosol particle per droplet;
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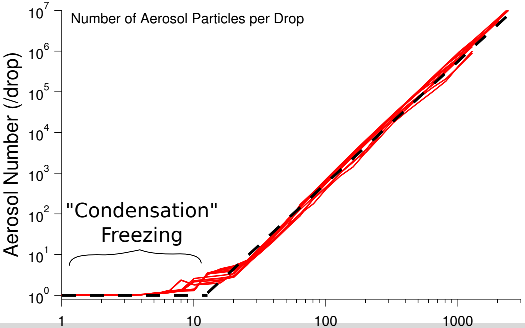

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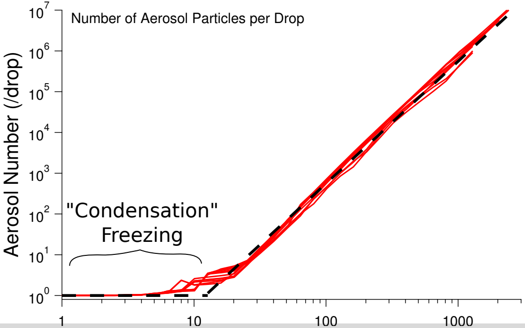

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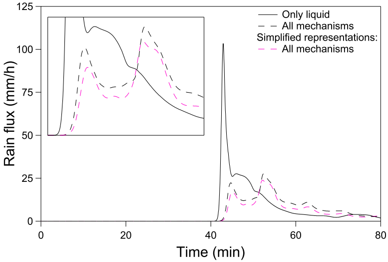
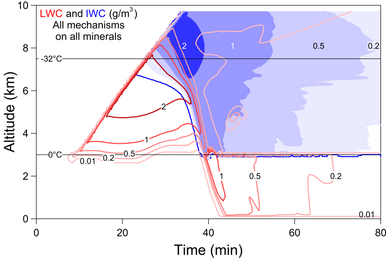
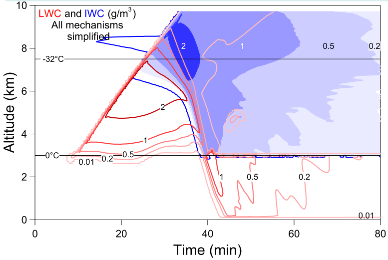


Results

- respective impact of the different mechanisms remain the same;
- dynamics changes because of surface computation modifications;
- minimal impact of the aerosol type simplification.





Descam

- Extension of the model for more detailed ice nucleation computation through implementation of INAS densities algorithms: $\Rightarrow$ Aerosol particles' size and type can be taken into account.
- Feldspar most ice active mineral and most impact on the cloud development.
- Immersion freezing most important mechanism for this dynamical framework, even at rather high altitudes, where homogeneous nucleation is active.
- Simplification of the ice nucleation algorithms to prepare for more complex dynamics.

Conclusion

- **Development of a new setup** for simultaneous investigation of deposition nucleation on multiple identified particles;
- **First experimental comparison** of immersion freezing/deposition nucleation efficiency *on the same identified* aerosol particles:
  $\Rightarrow$ **Good ice nuclei for immersion freezing seem to be good ice nuclei for deposition nucleation**.

- Improvement of heterogeneous nucleation computation in DESCAM for **highly detailed studies of ice nucleation**: state of the art parametrizations with INAS density representation from the literature and from this thesis' experiments;
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- Combination of the experiments with ESEM investigation of the samples, particularly on identified efficient sites.

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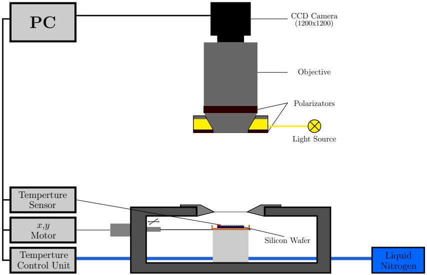
Thank You for Your Attention!

Cold Stage exp

Material

- Linkam motorized Cold Stage MDBCS196
- Up to 2000 droplets printed on a silicon wafer
- Individual freezing of droplets detected by depolarization



Limitations

Fine control of stage temperature, but:

- little knowledge on ambient temperature;
- no control over the ambient humidity;
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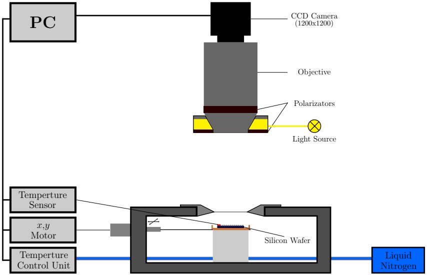

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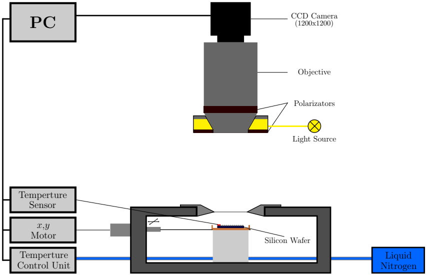

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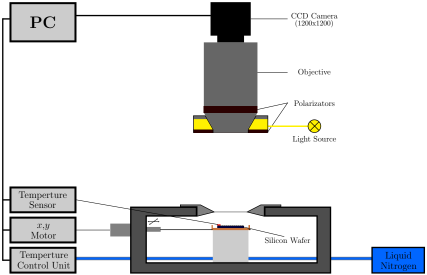
Fine control of stage temperature, but:

- little knowledge on ambient temperature;
- no control over the ambient humidity;
- RH control complicated due to large open volume of the chamber.

Cold Stage exp

Material

- Linkam motorized Cold Stage MDBCS196
- Up to 2000 droplets printed on a silicon wafer
- Individual freezing of droplets detected by depolarization



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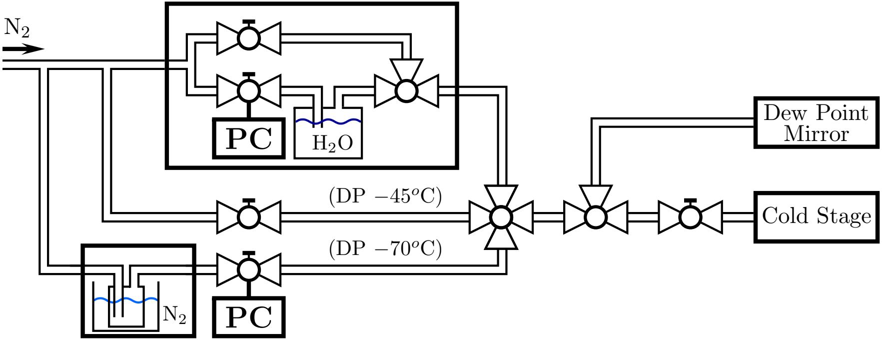
Relative Humidity System

Performance

- Humidity controlled for $T_{DP} \in [-44; 3°C]$
- T_{DP} down to $-70°C$

Limitations

- P_w approximated by fit
- Inflow limited to 150 ml/min
- No local measure of RH



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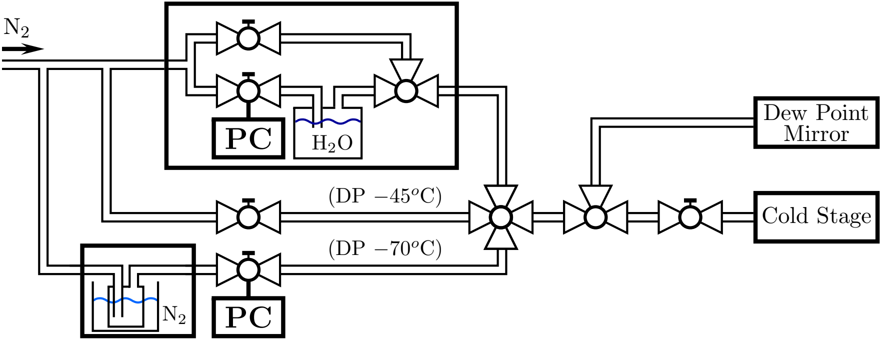

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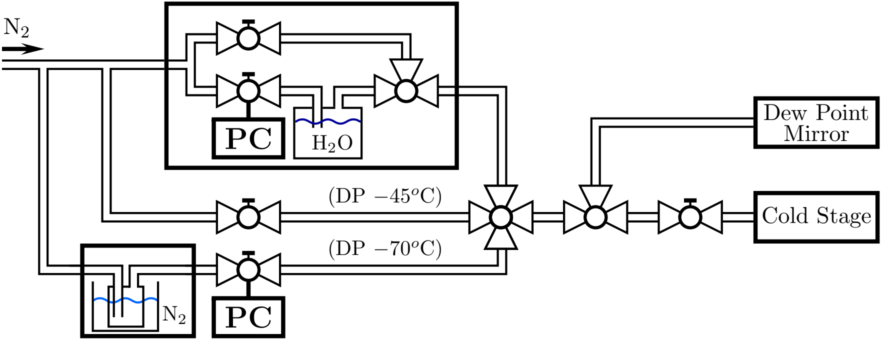

Immersion freezing

Ice nuclei identification

For consecutive freezing experiments, a good correlation in freezing rank indicates heterogeneous nucleation (Peckhaus *et al.* 2016).

- Clear correlation for suspension **B**: heterogeneous nucleation;
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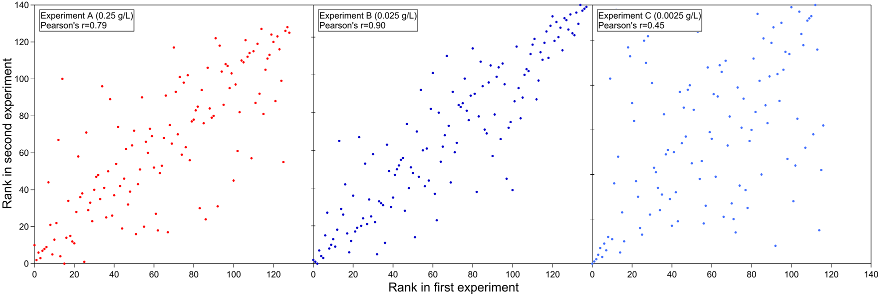

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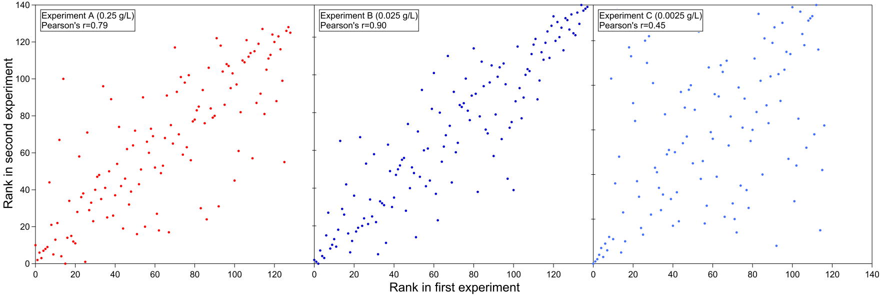

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Ice Nucleating Active Sites (INAS) densities

Mean of description of experimental data defined as:

$$n_{s,imm}(T) = -\frac{1}{A_{aer}} \cdot \ln\left(1 - f_{IN}(T)\right)$$

with $f_{IN}(T)$ the frozen fraction and A_{aer} the surface of aerosol particle per droplet.

Parametrization

Reduced range of validity (low concentrations of aerosol particles in suspension).
Data fit based on the double exponential fit function from Hiranuma et al. (2015) for illite NX:

$$n_s(T) = \exp(10.3 \cdot \exp[-\exp(0.345 \cdot (T + 21.2))] + 6.05)$$

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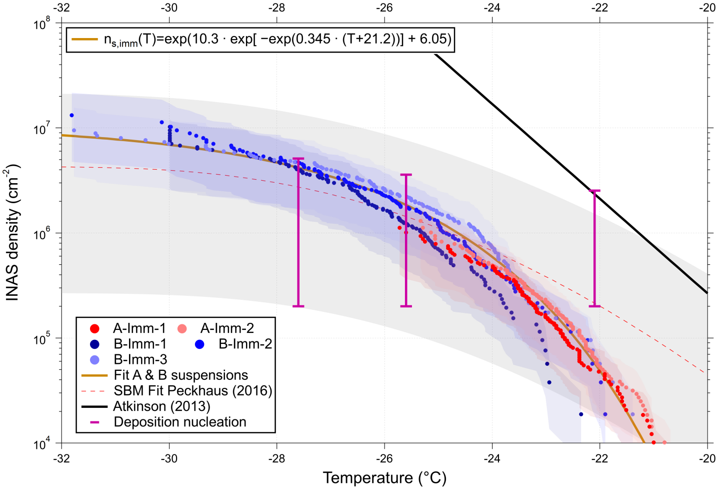

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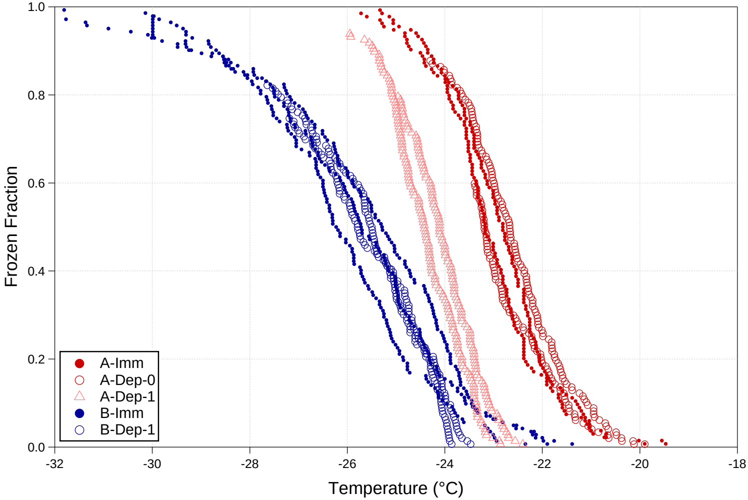

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Summary of deposition nucleation experiments

Exp No.	CS Frost Point	Crystals
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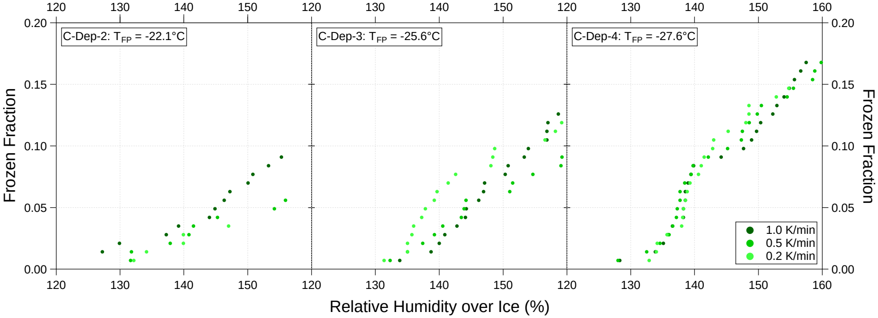

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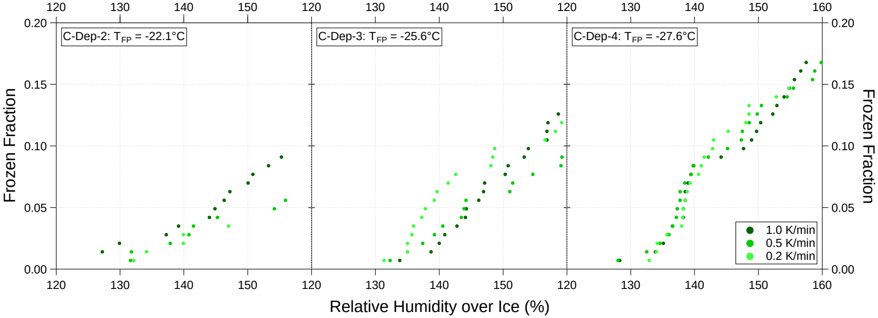

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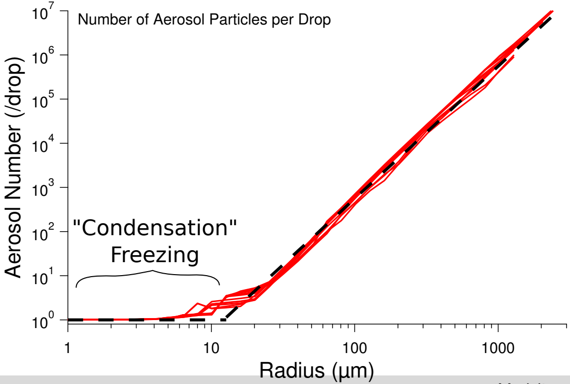

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$$\begin{cases} n_{IN,imm}(j,kt) = (\mathcal{N}_d(j,kt) + \mathcal{N}_i(j,kt)) \cdot (1 - \alpha_{imm}(j,kt)) & \text{if} \quad j(r_d) \\ n^{\dagger}_{IN,imm}(j) = \qquad (N_d(j) + N_i(j)) \cdot (1 - \prod_{kt} \alpha^{\dagger}_{imm}(j,kt)) & \text{else} \end{cases}$$

where j and kt correspond to the size bin and considered aerosol type respectiv
nucleation ratio, which is defined as:

$$\begin{cases} \alpha_{imm}(j,kt) = \exp\left(-n_{s,imm}(T,kt) \cdot \frac{\mathcal{S}_d(j,kt) + \mathcal{S}_i(j,kt)}{\mathcal{N}_d(j,kt) + \mathcal{N}_i(j,kt)}\right) & \text{if} \quad j(r_d \\ \alpha^{\dagger}_{imm}(j,kt) = \exp\left(-n_{s,imm}(T,kt) \cdot \frac{\mathcal{S}_d(j,kt) + \mathcal{S}_i(j,kt)}{N_d(j) + N_i(j)}\right) & \text{else} \end{cases}$$

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This yields the number of new ice crystals:

$$\mathrm{d}N_{i,imm}(j) = \begin{cases} \max\left[0, \sum_{kt} n_{IN,imm}(j,kt) - N_i(j)\right] & \text{if} \quad j(r_d) \leq 1 \\ \max\left[0, n^{\dagger}_{IN,imm}(j) - N_i(j)\right] & \text{else} \end{cases}$$

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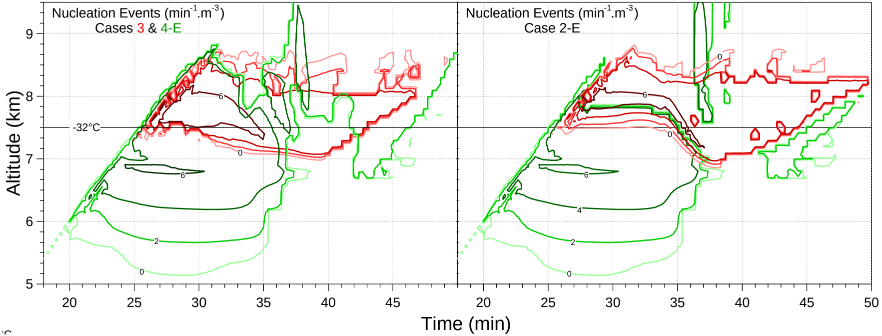
Condensation and immersion freezing: same parametrization,
algorithms.
Condensation freezing ⇒ only one aerosol particle per drople
Immersion freezing ⇒ uniform mix of aerosol particles in the

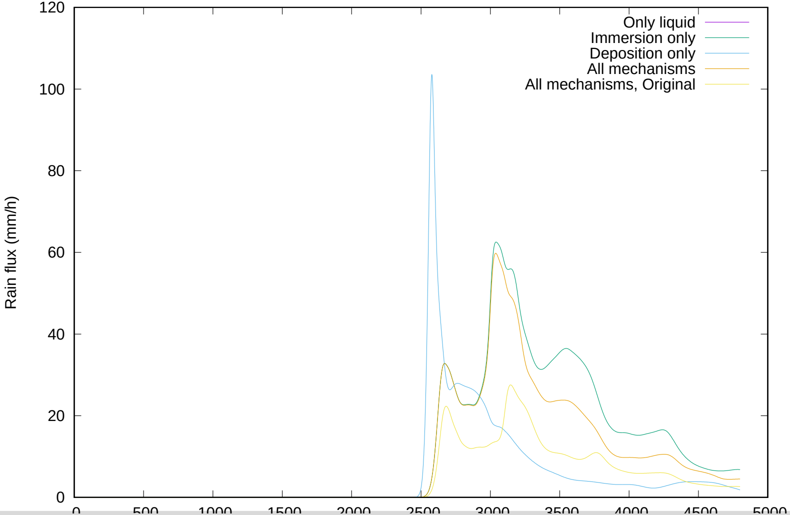


Because of the competition between the two mechanisms:

- reduced homogeneous ice nucleation rate between 7.5 and 7.8 km;
- immersion freezing inactive above 7.8 km.

Less reduction of homogeneous freezing than in preliminary study.

Ullrich, 2017 parametrization



Mineral concentration c0/100