Measurement campaign FROST
-
Results of LACIS ice experiments

Dennis Niedermeier, Susan Hartmann, the FROST-Team,
and Frank Stratmann

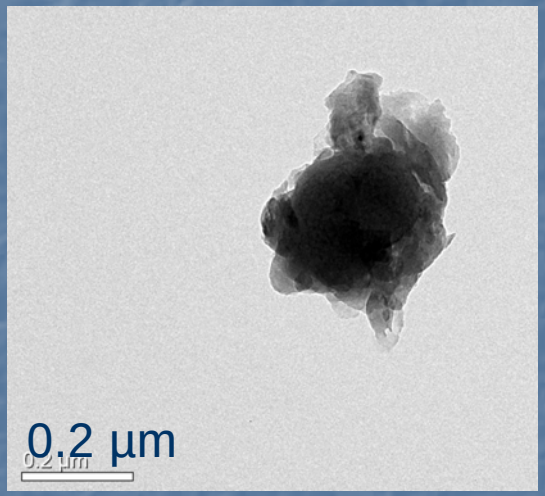
Virtual Institute
Aerosol Cloud Interaction

FROST

- FROST: FReezing and activation abilities Of coated and uncoated mineral duST particles

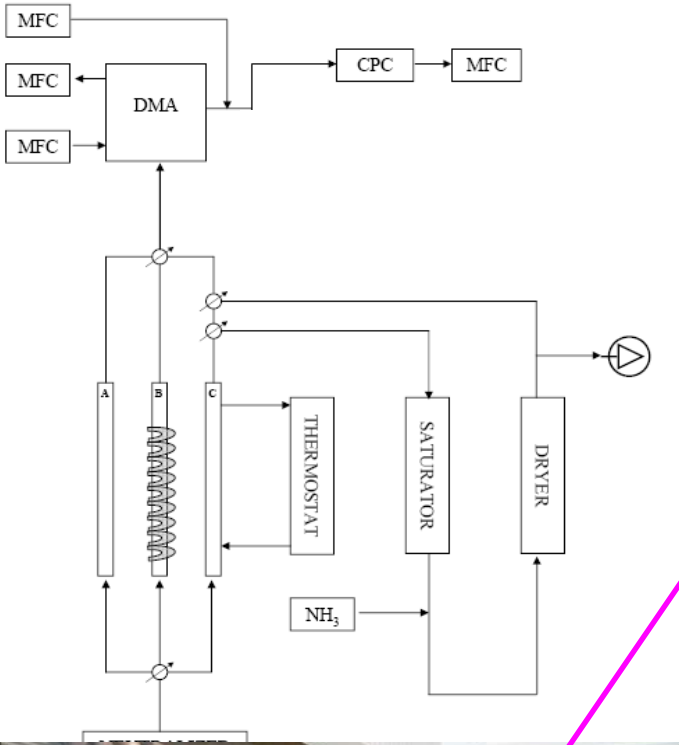
  - Freezing with LACIS (IfT)
  - Hygroscopic growth with HH-TDMA (IfT)
  - Activation with CCNC (IfT and University of Vienna)
  - Chemical composition with AMS (University of Mainz, FZ Jülich, and IfT)
  - Particle morphology through TEM-Analysis (MPI for Polymer Research, Mainz)

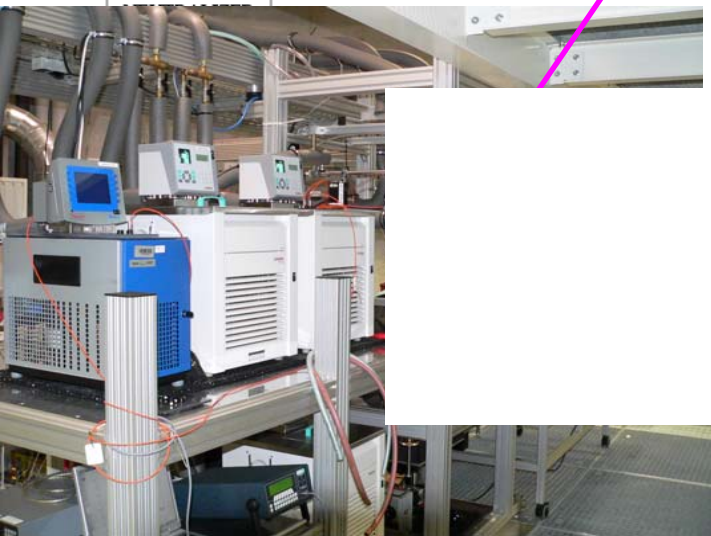
- First freezing measurements with LACIS
- Influence of different coatings on freezing behavior of dust particles
  - Arizona Test Dust (ATD) particles D_{mob} = 300nm
  - Various coatings: Uncoated, $C_4H_6O_4$, H_2SO_4 (50°C and 70°C), $(NH_4)_2SO_4$



ATD: SiO_2, Al_2O_3, ...

FROST - LACIS measurement setup





AIR

VACUUM EXHAUST

$T_{d,in}$ = -7°C

OPC

$T_{w,1}$ = 20°C

$T_{w,2}$ = 0°C

$T_{w,3}$ = 0°C

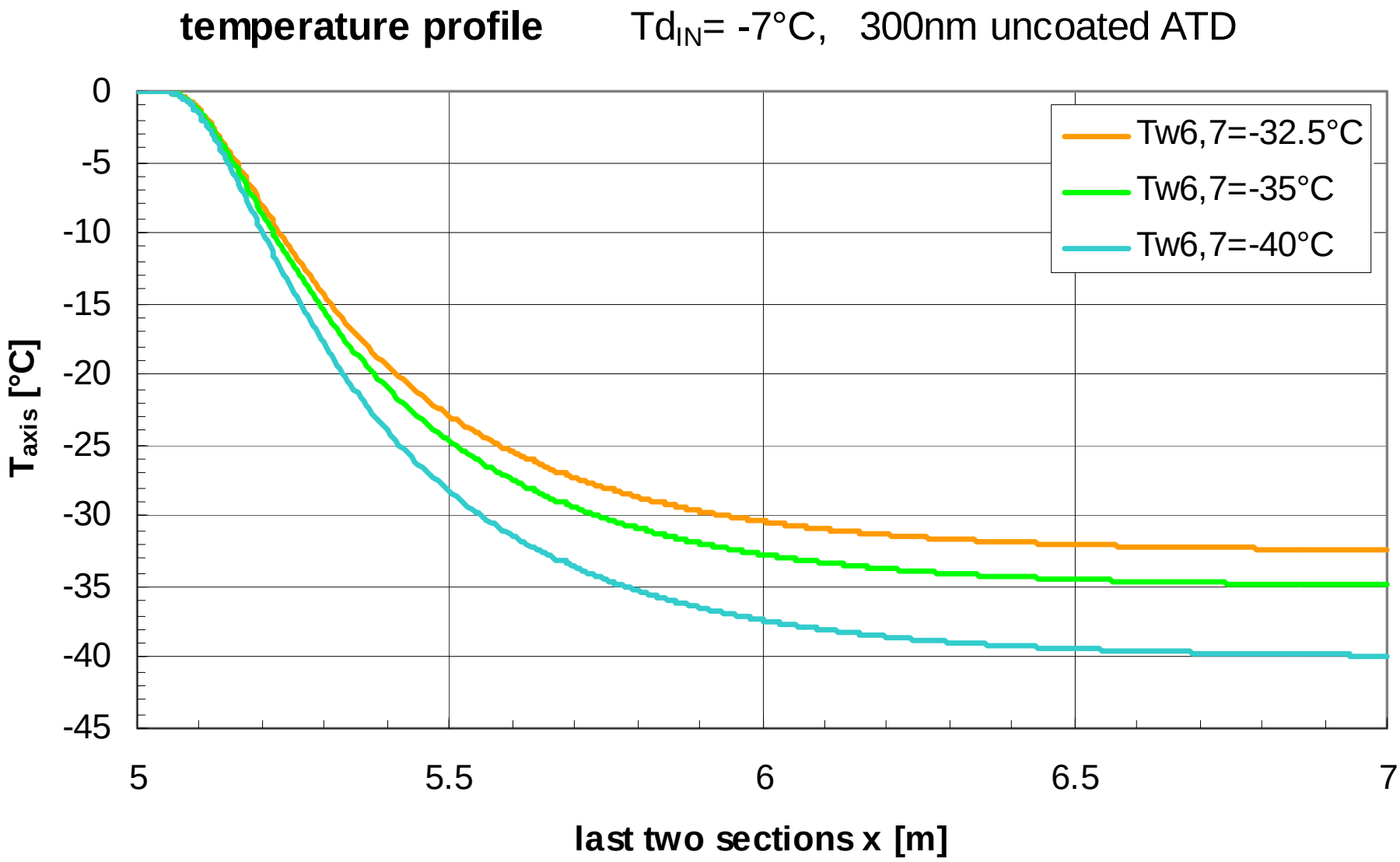
$T_{w,4}$ = 0°C

$T_{w,5}$ = 0°C

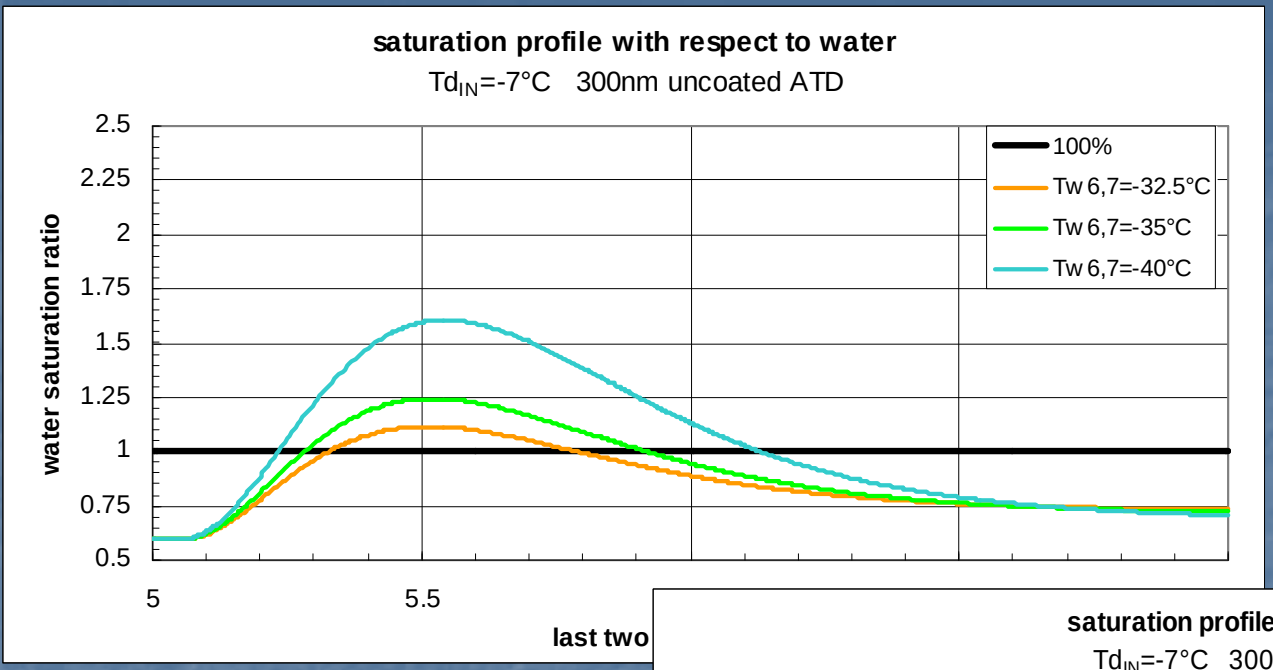
$T_{w,6}$ = 0°C, variable

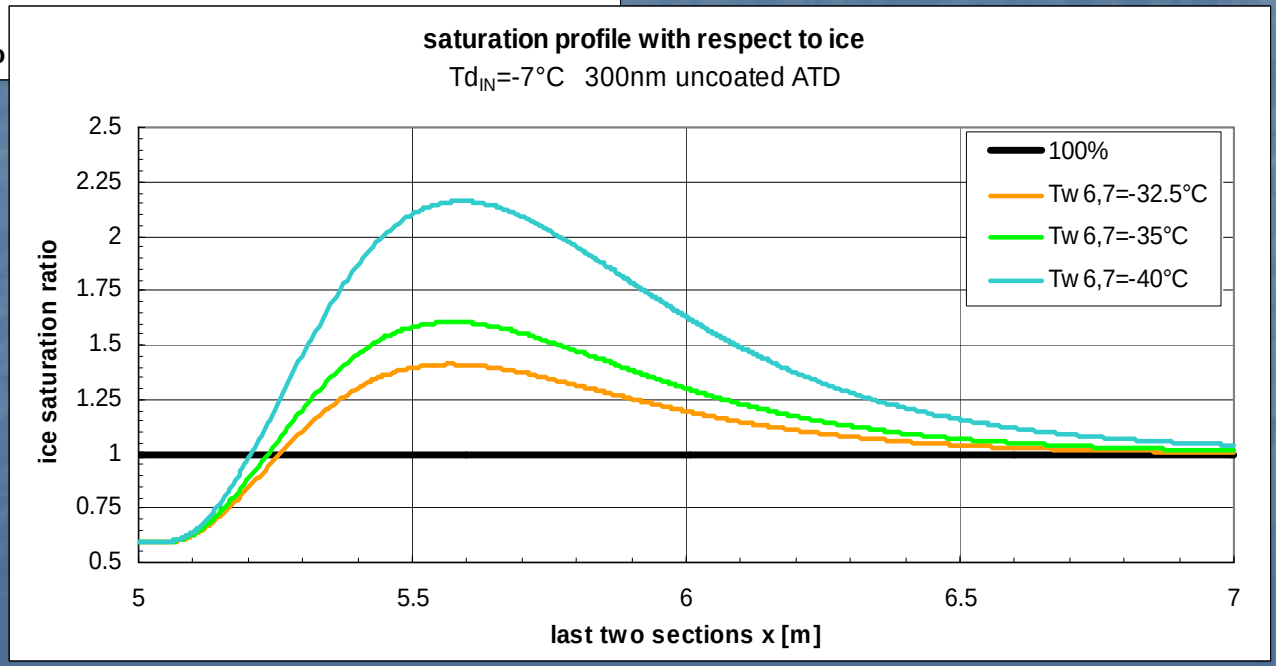
$T_{w,7}$ = variable

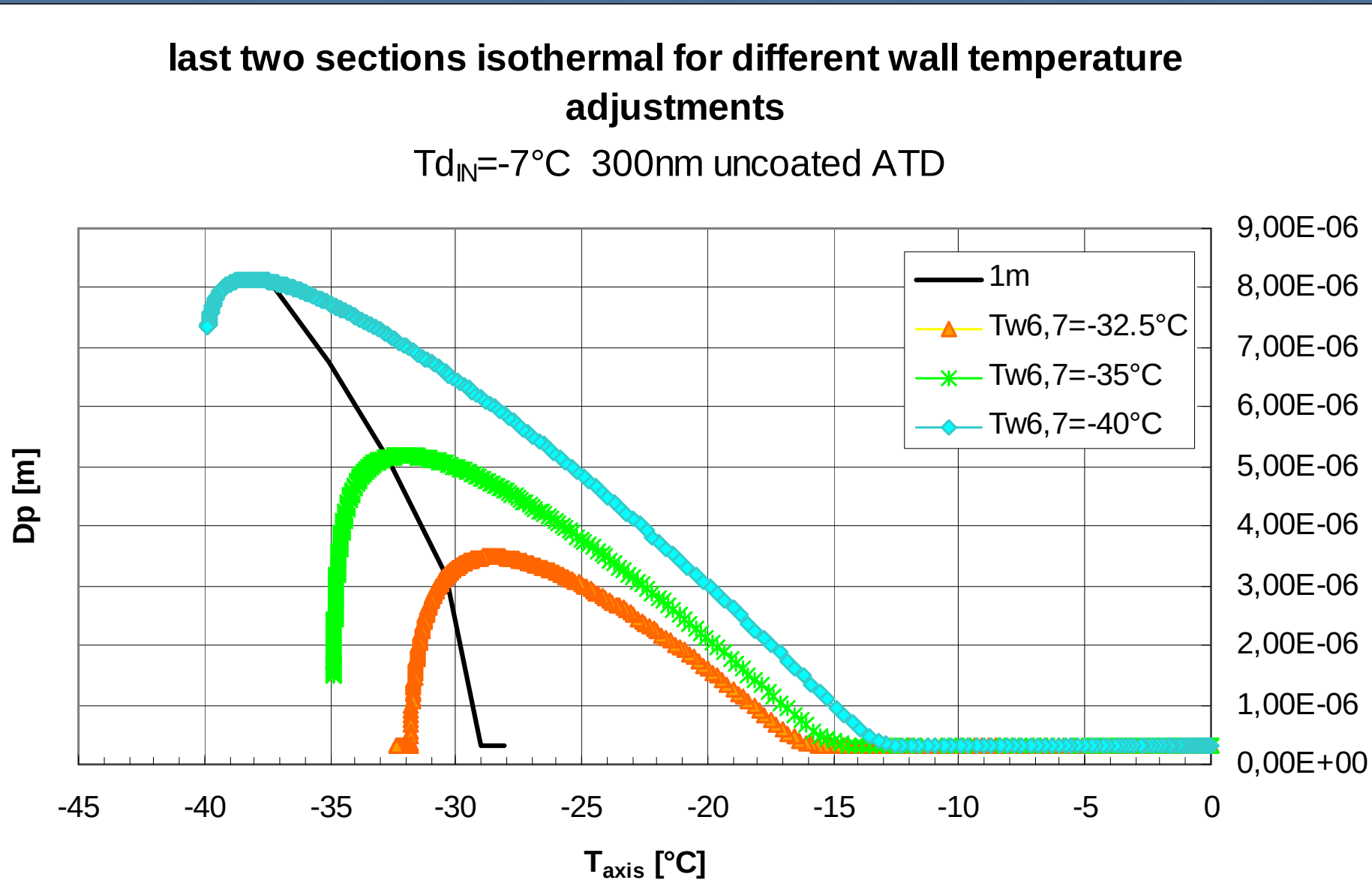
WELAS


temperature profile
Td$_{IN}$= -7°C, 300nm uncoated ATD
Tw6,7=-32.5°C
Tw6,7=-35°C
Tw6,7=-40°C
T$_{axis}$ [°C]
0
-5
-10
-15
-20
-25
-30
-35
-40
-45
5
5.5
6
6.5
7
last two sections x [m]

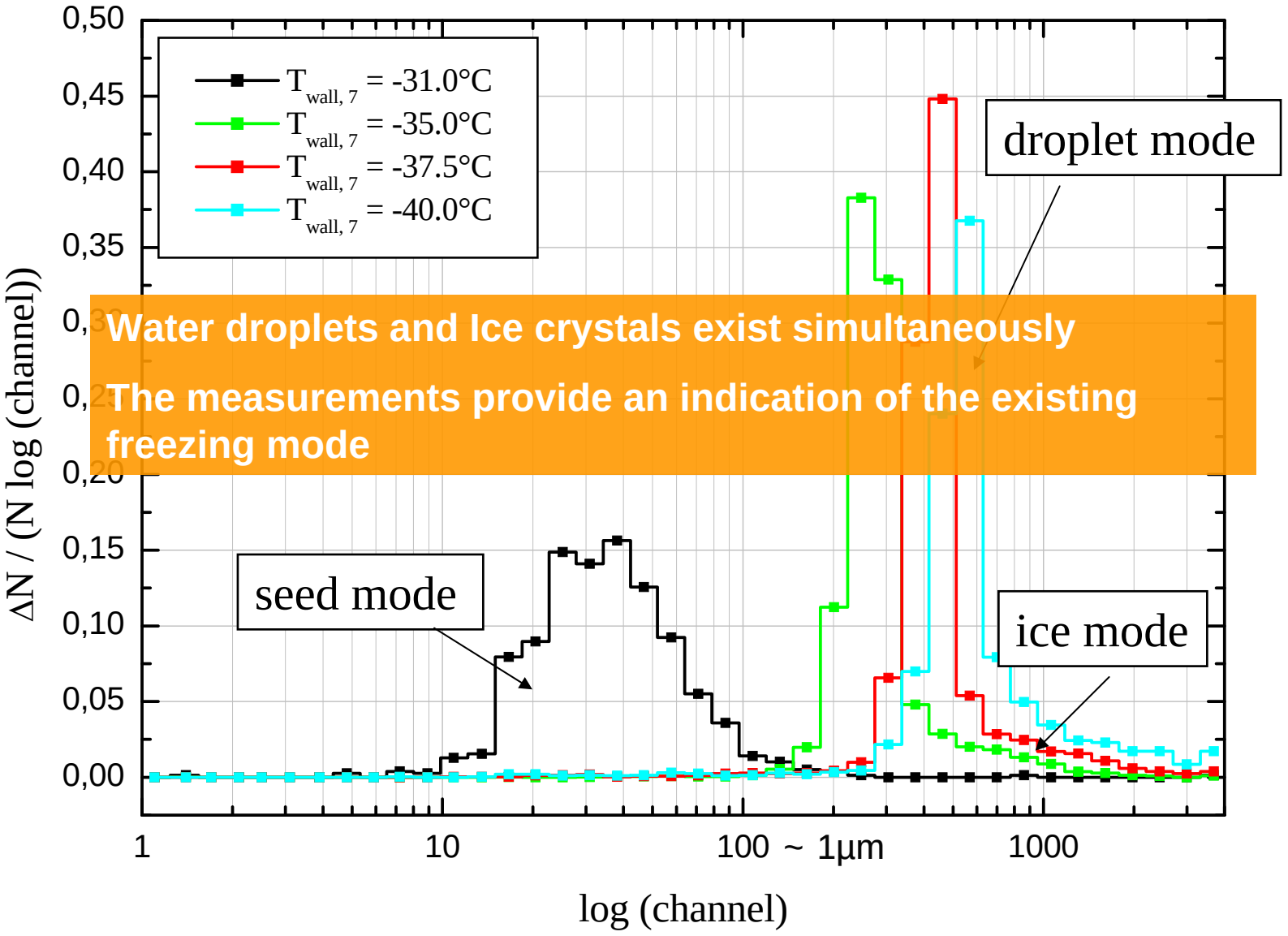
FROST - LACIS measurement setup






last two sections isothermal for different wall temperature adjustments
Td$_{IN}$=-7°C 300nm uncoated ATD
Dp [m]
9,00E-06
8,00E-06
7,00E-06
6,00E-06
5,00E-06
4,00E-06
3,00E-06
2,00E-06
1,00E-06
0,00E+00
1m
Tw6,7=-32.5°C
Tw6,7=-35°C
Tw6,7=-40°C
-45
-40
-35
-30
-25
-20
-15
-10
-5
0
T$_{axis}$ [°C]

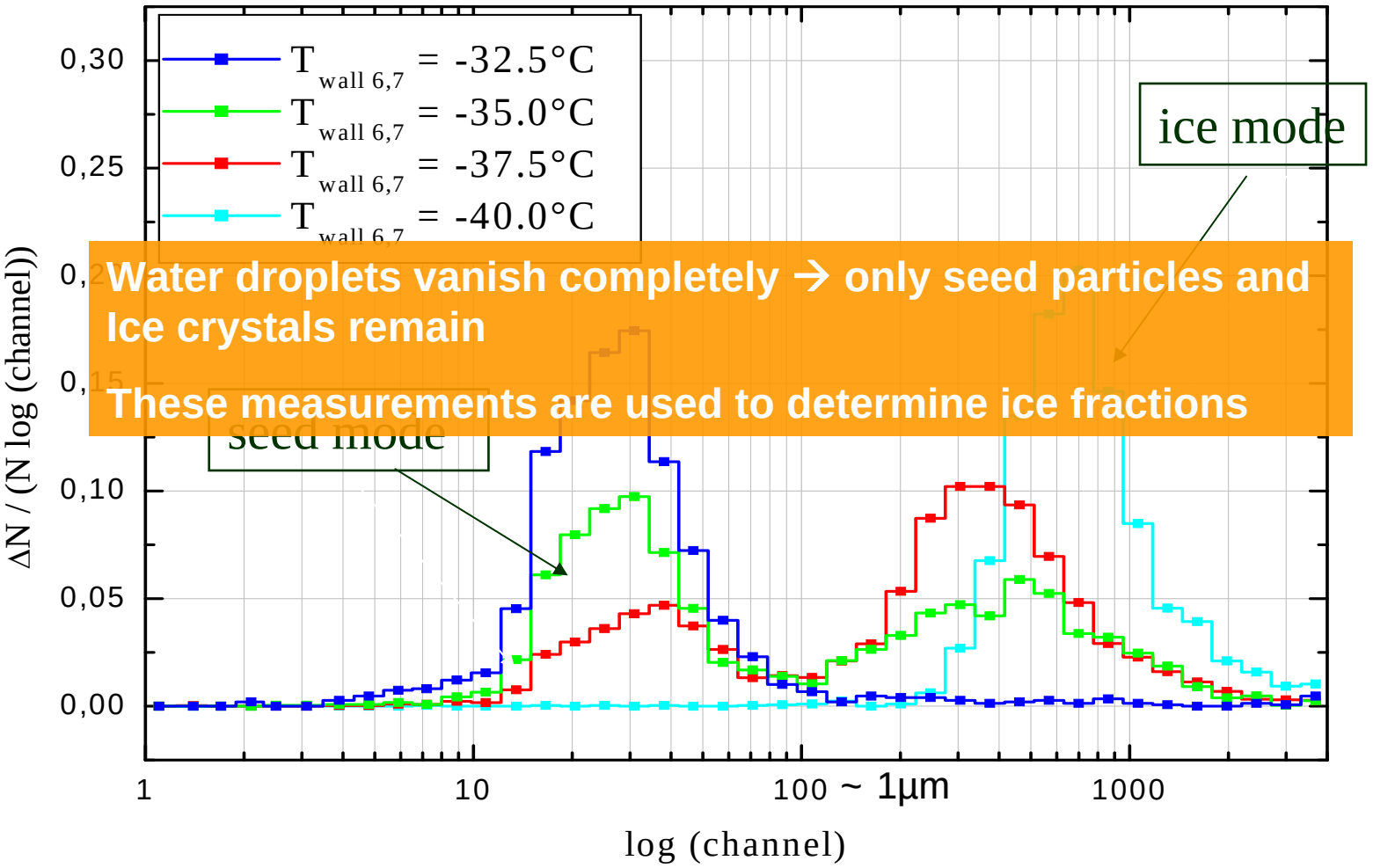
Measurement with last one meter section; 300 nm uncoated ATD



Water droplets and Ice crystals exist simultaneously

The measurements provide an indication of the existing freezing mode

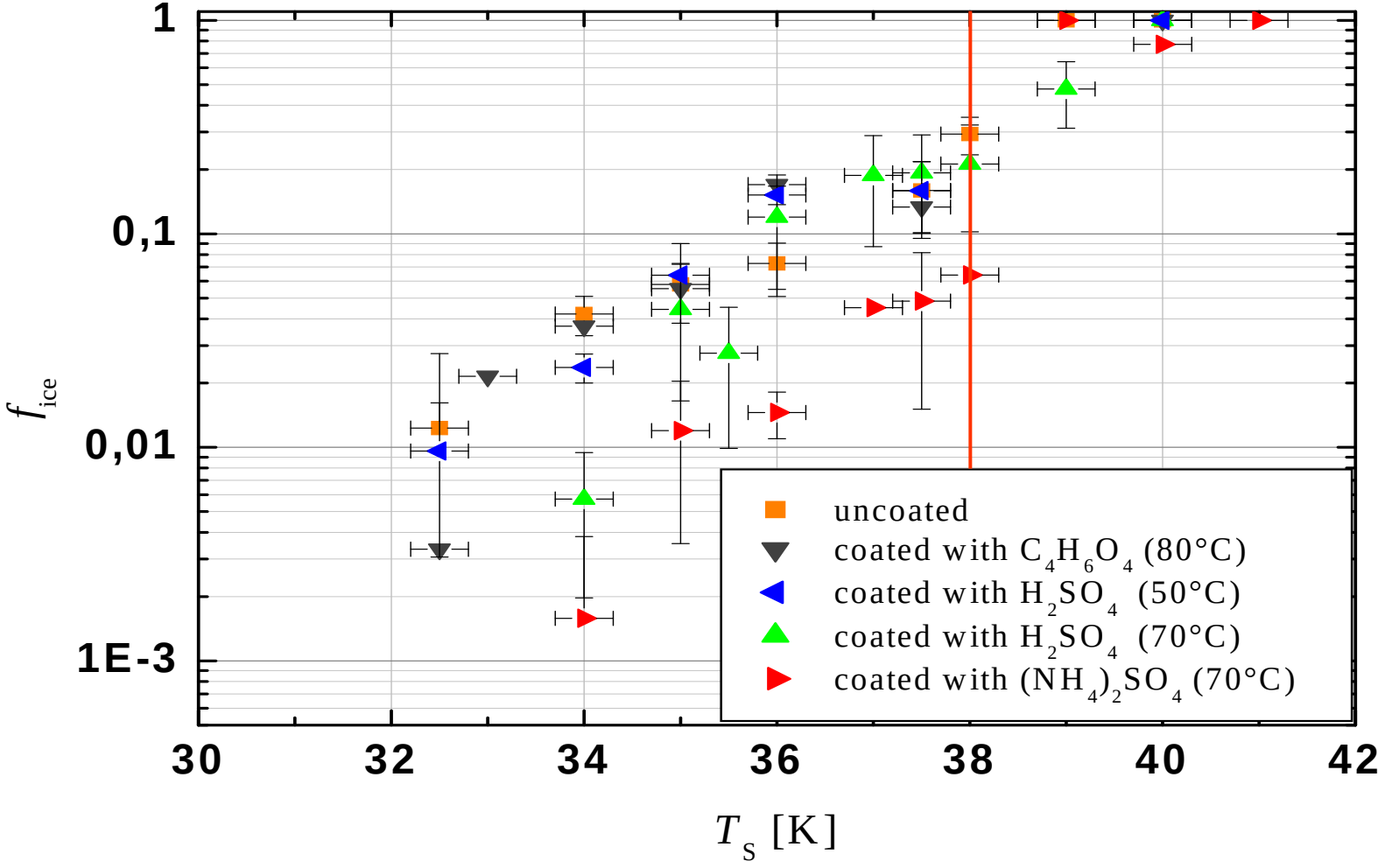
Measurements with last two sections; 300 nm uncoated ATD particles

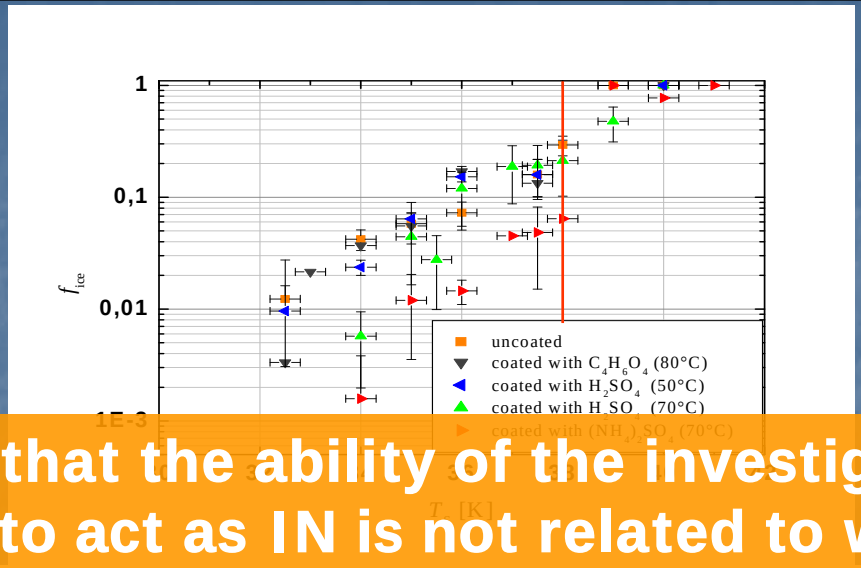


Water droplets vanish completely → only seed particles and Ice crystals remain

These measurements are used to determine ice fractions

Ice fraction for all types of particles → D_{mob} = 300nm

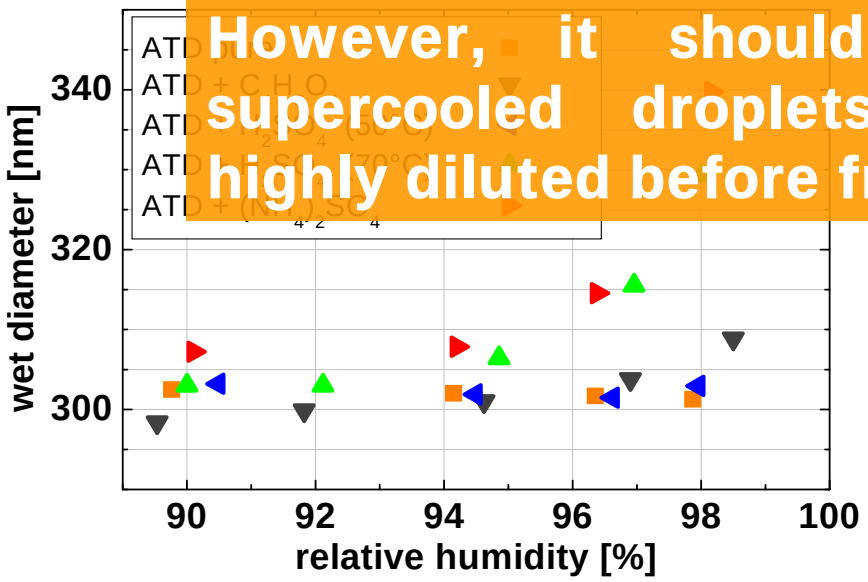




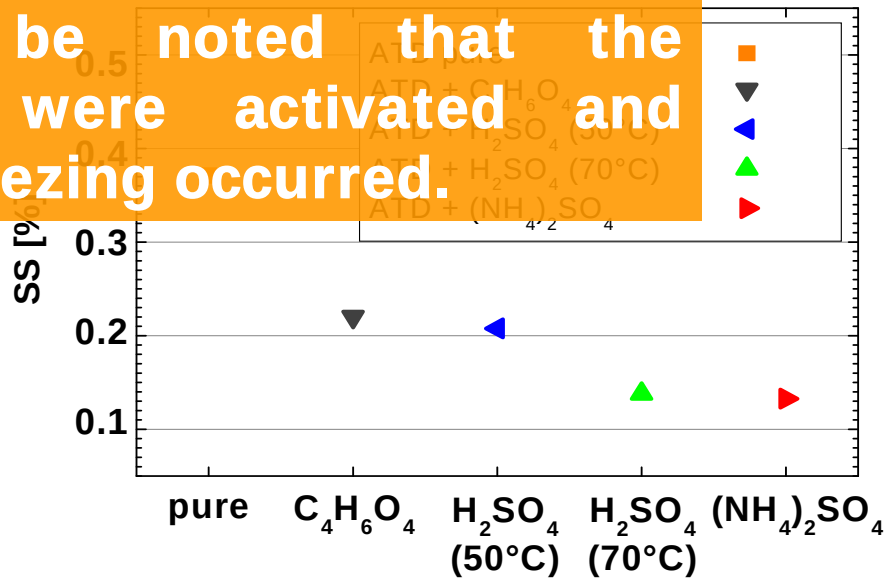
It seems that the ability of the investigated particles to act as IN is not related to water activity.

However, it should be noted that the supercooled droplets were activated and highly diluted before freezing occurred.

HH-TDMA



CCNC



A fit procedure, based on a simplified CNT type nucleation rate expression was performed

Two Assumptions: 1. constant nucleation rate (constant T_s)
2. stochastic process

$$f_{\mathrm{ice}} = 1 - \exp\left(-\omega_{\mathrm{het}} t\right)$$

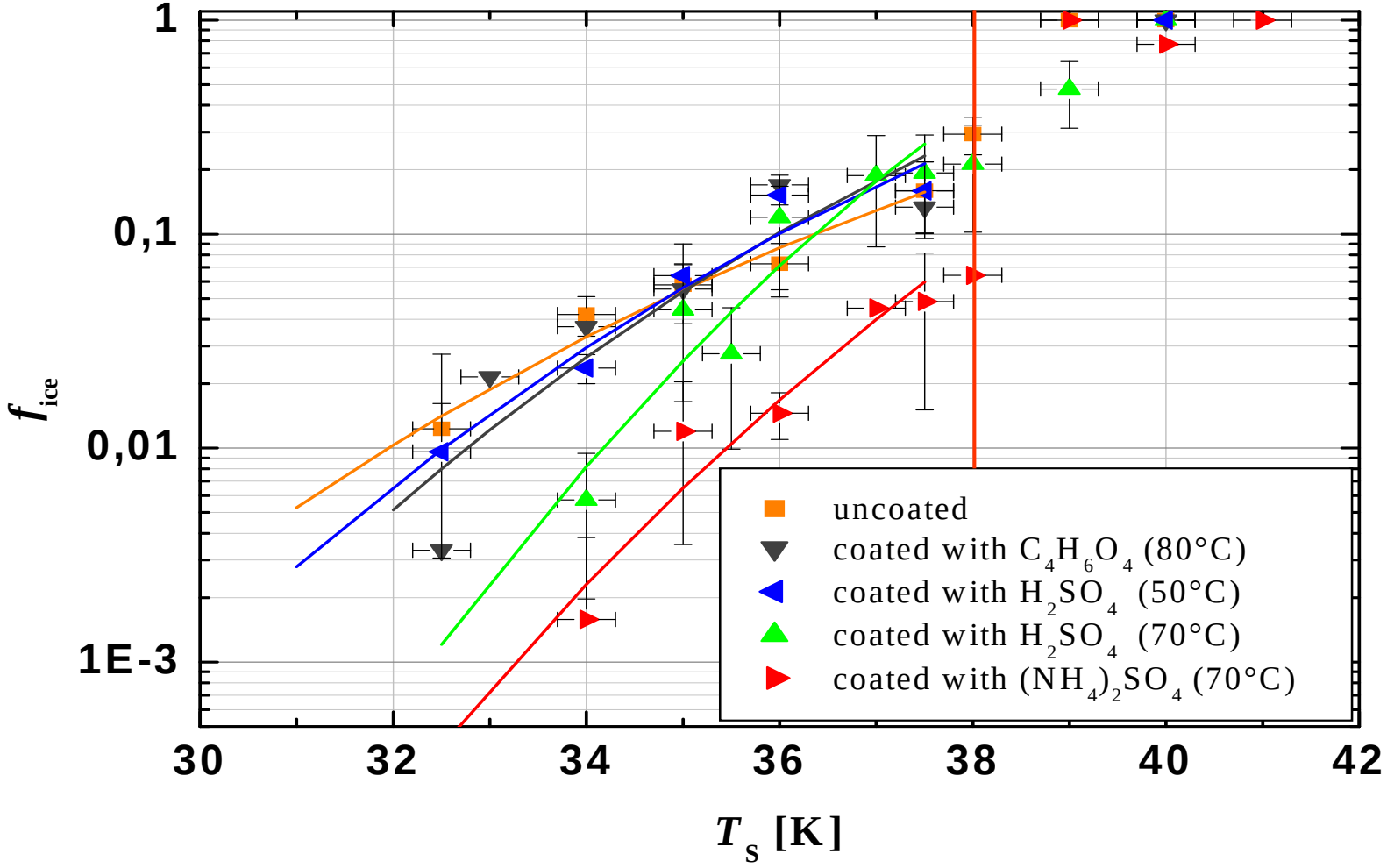
$$\omega_{\mathrm{het}} = \alpha \cdot \exp\left(-\frac{\beta}{T_s^2}\right)$$

Total surface and Kinetics

Thermodynamics

$$f_{\mathrm{ice}} = 1 - \exp\left(-\alpha \cdot \exp\left(-\frac{\beta}{T_s^2}\right) t\right)$$

Ice fraction for all types of particles → including fitted curves



- Influence of different coatings on freezing behavior of dust particles (D_{mob} = 300nm) was investigated
- Uncoated particles and those with $C_4H_6O_4$ or thin H_2SO_4 coatings show similar freezing behavior
- Particles with thick H_2SO_4 or with $(NH_4)_2SO_4$ coatings start to serve as IN at lower supercooling temperature
- A CNT-type rate expression was successfully applied to parameterize measured ice fractions
- Publications in preparation:
  - LACIS for ice nucleation studies (IfT)
  - Immersion freezing of coated/uncoated ATD
  - Determination of coating with AMS (Mainz)

- FROST II
  - Freezing with CFDC (CSU), PINC (ETH Zürich), FINCH (Uni Frankfurt), and LACIS (IfT)
  - Hygroscopic growth with H-TDMA (FZJ)
  - Activation with CCNC (IfT)
  - Chemical composition with AMS (University of Mainz) and ATOFMS (ETH Zürich)
  - Particle morphology (IfT, MPI Mainz, ...)

FROST II (March/April 2009) and afterwards:

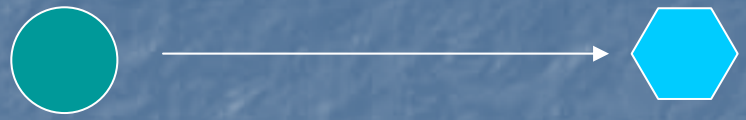
- Influence of surface changes on IN ability → Do surface treatments (coatings, heating, denuding) change the IN ability of ATD particles ? *(YES)*
- Influence of particles size on IN ability →Freezing experiments with larger monodisperse ATD particles (~ 1µm) *(postponed)*
- Using new optical device to determine ice fractions (TOPF-ICE and SID) *(in progress)*

Treatment	Ice fraction for T_s = 35K
Pure ATD	5.83% (+/-3.33%)
Pure ATD + TD	3.84% (+/-2.41%)
ATD + H_2SO_4 45°C	5.12% (+/-2.98%)
ATD + H_2SO_4 45°C + TD	0.33% (+/-0.22%)
ATD + H_2SO_4 70°C	1.23% (+/-0.72%)
ATD + H_2SO_4 70°C + TD	0.06% (+/-0.03%)
ATD + H_2SO_4 70°C + H_2O bath	0.28% (+/-0.17%)
ATD + H_2SO_4 70°C + H_2O bath + TD	0.42% (+/-0.24%)
ATD + H_2SO_4 70°C + H_2O bath + NH_3	0.27% (+/-0.16%)
ATD + H_2SO_4 70°C + H_2O bath + NH_3 + TD	0.40% (+/-0.21%)

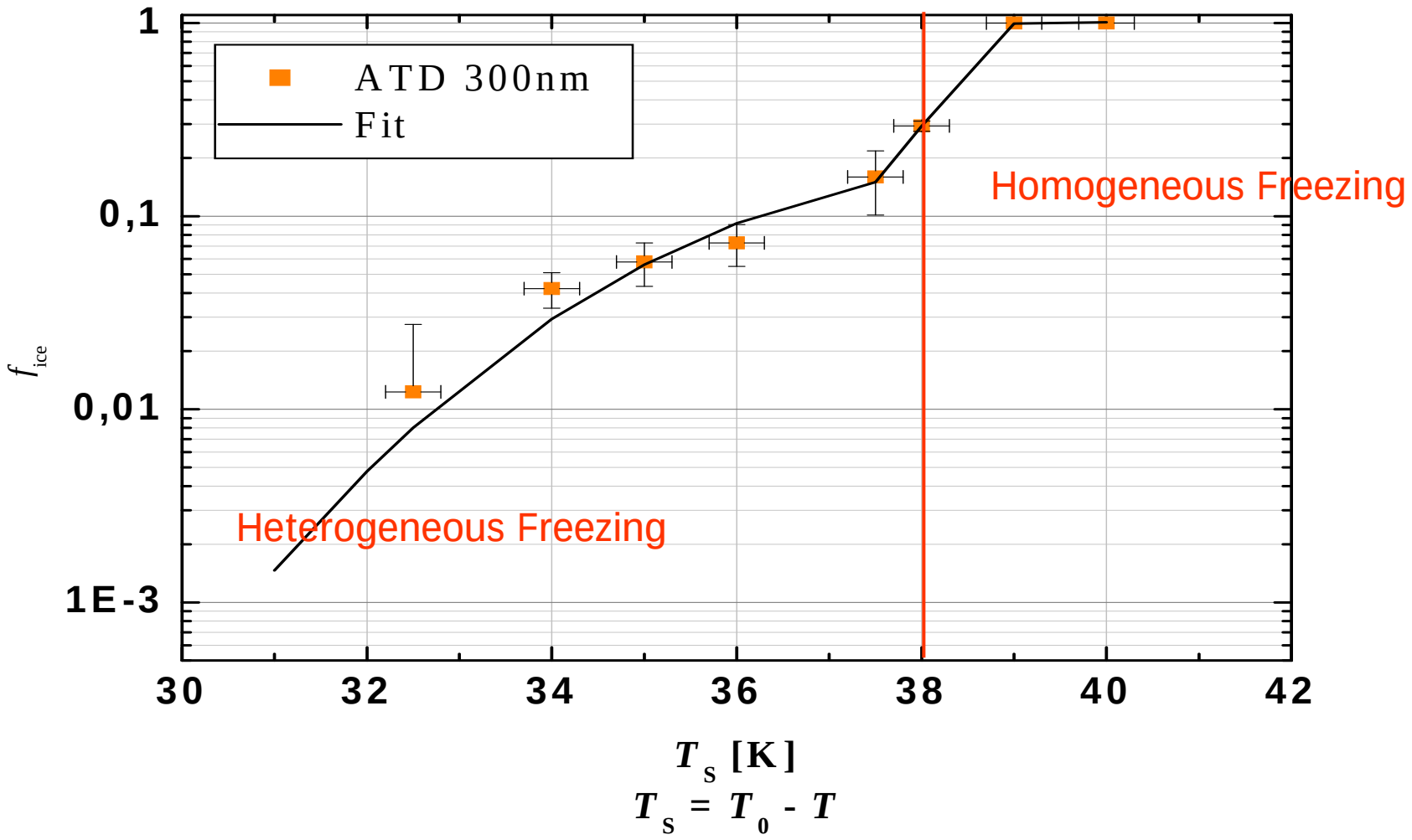
THE END

- Which freezing modes occur?

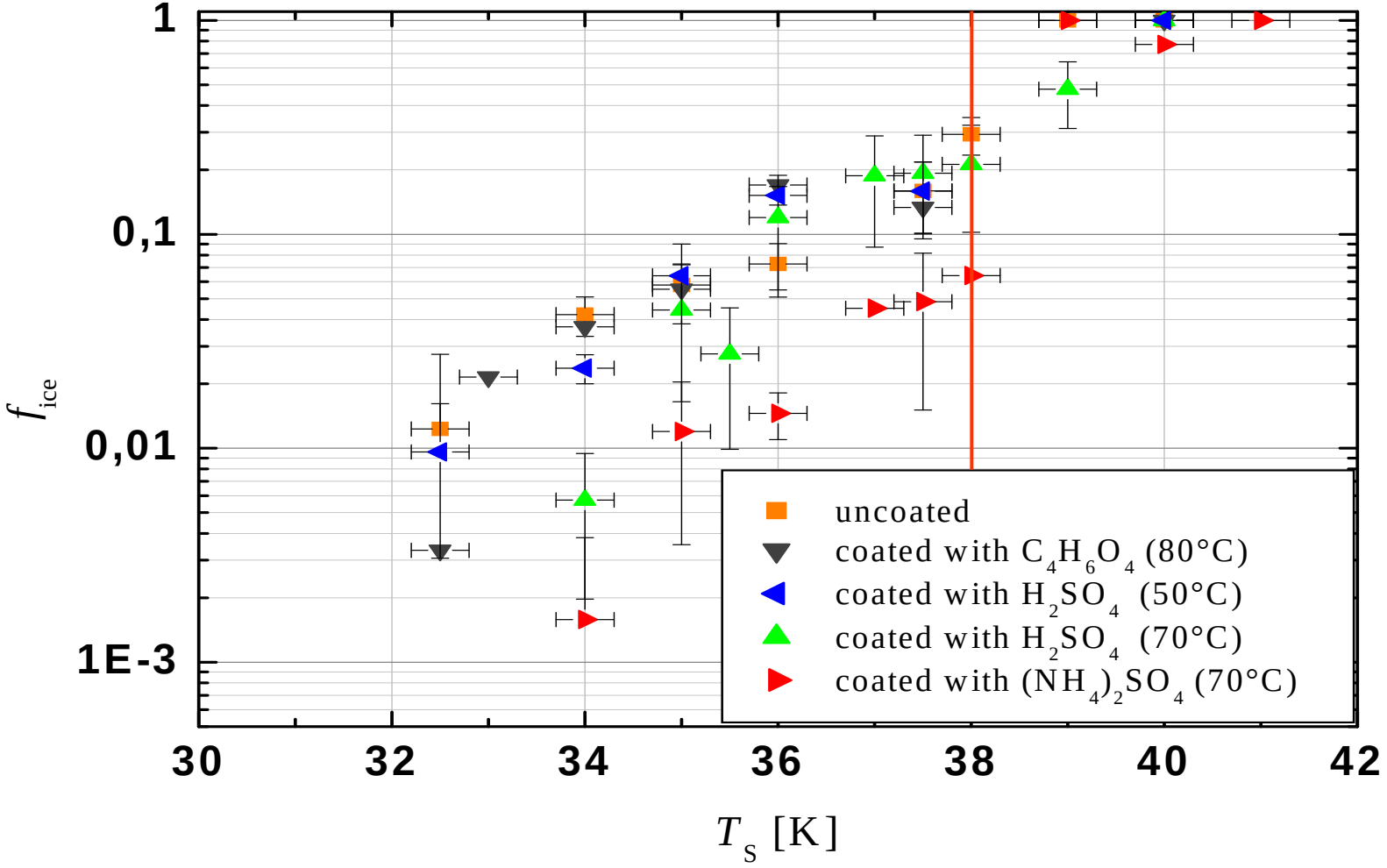
Homogenous freezing ?



Ice fractions for 300 nm uncoated ATD particles



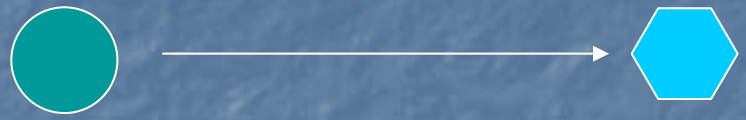
Ice fraction for all types of particles → D_{mob} = 300nm



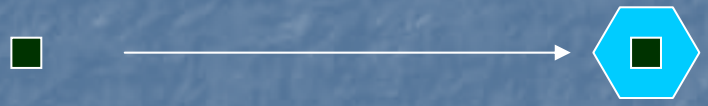
- Which freezing modes occur?

Homogenous freezing

→ occurs for $T_s \geq 38K$

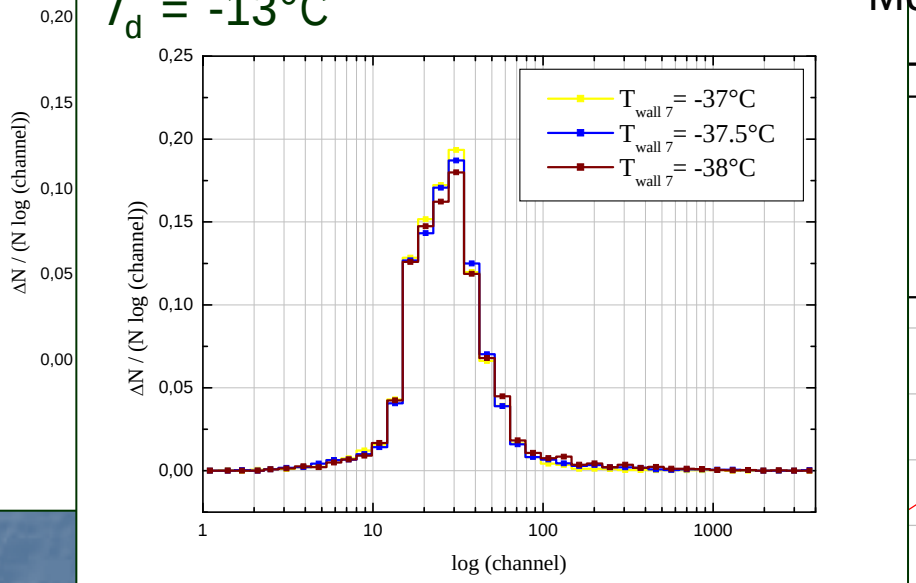


Deposition freezing ?

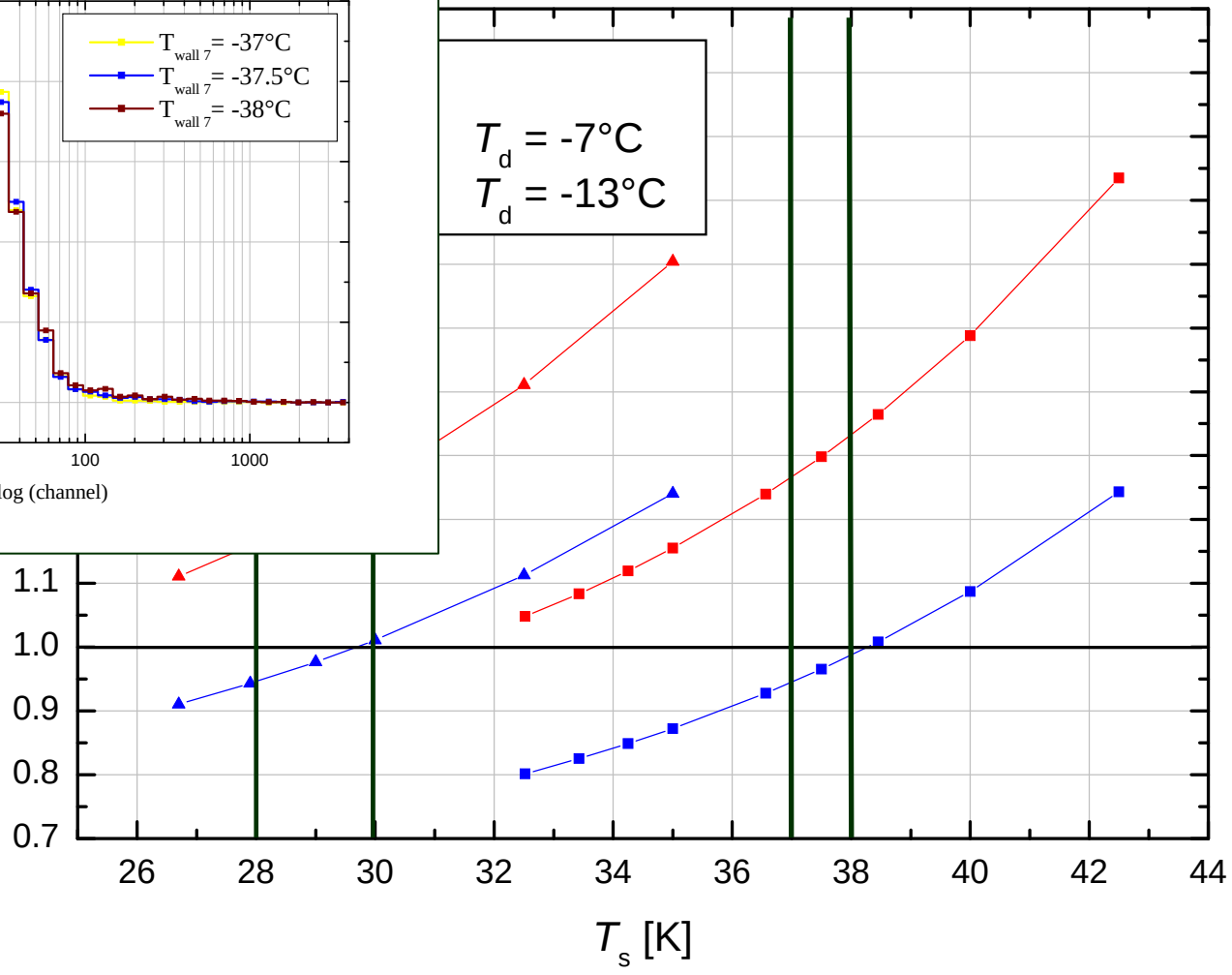


T_d = -7°C

T_d = -13°C

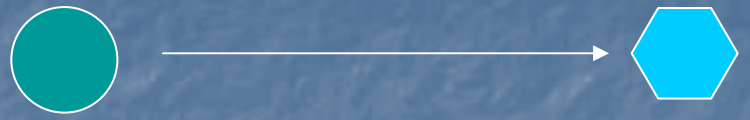


Model Calculations



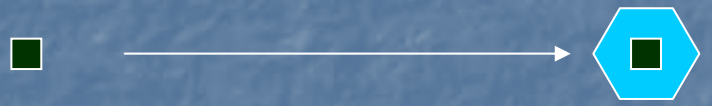
Which freezing modes occur?

Homogenous freezing



→ occurs for $T_s \geq 38K$

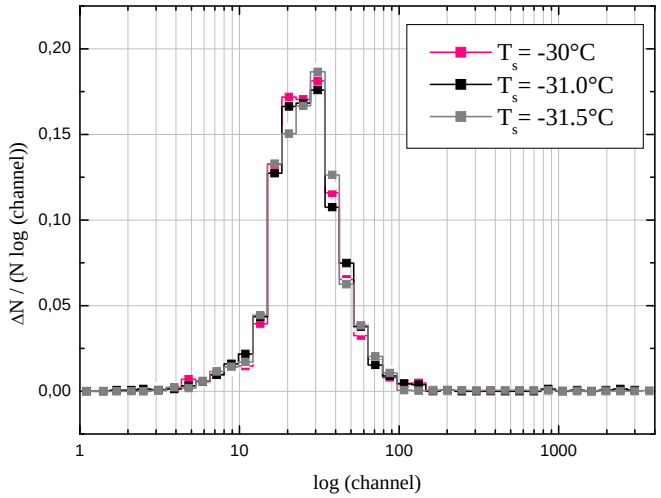
~~Deposition freezing~~



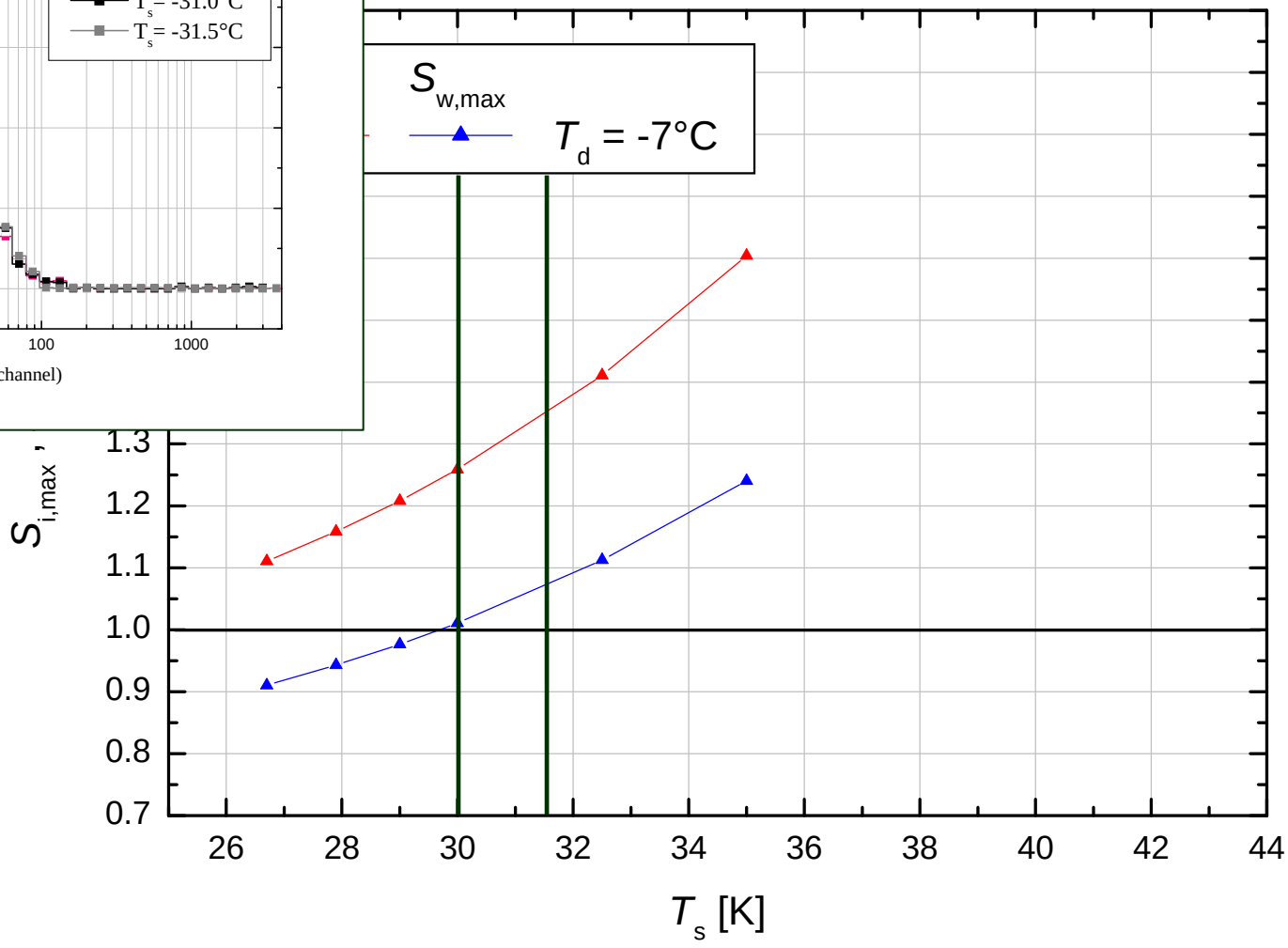
→ excludable because of extra performed measurements

Evaporation freezing ?

T_d = -7°C

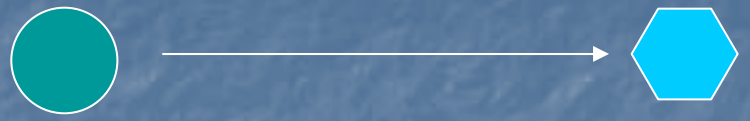


Model Calculations



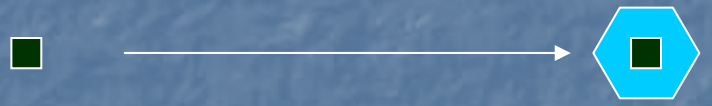
Which freezing modes occur?

Homogenous freezing



→ occurs for $T_s \geq 38K$

~~Deposition freezing~~

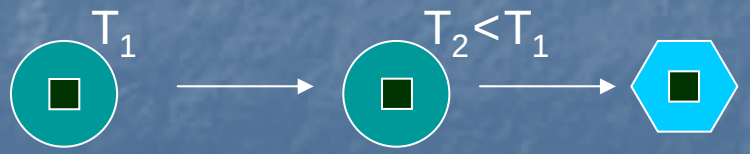


→ excludable because of extra performed measurements

~~Evaporation freezing~~

→ also excludable

Immersion freezing



→ occurring heterogeneous freezing mode