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# 1D and Super-Particle Aerosol-Cloud Modeling to Assess the Role of Cloud Conditions and Immersion Freezing Parameterizations on the INP Reservoir

ASR Atmospheric System Research

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## Motivation

High latitude mixed-phase clouds (MPCs) contribute significantly to the uncertainty in estimated equilibrium climate sensitivity. This uncertainty is primarily driven by deficient knowledge of cloud processes, which determine the supercooled liquid and ice fraction, and hence, the cloud's reflectivity on mid-to-large scales. To advance our understanding of the underlying cloud microphysical processes we employ a minimalistic 1D aerosol-cloud (1D AC) model and a super-particle model to prognostically evaluate the evolution of the ice-nucleating particle (INP) reservoir and resulting ice crystal (IC) number concentrations.

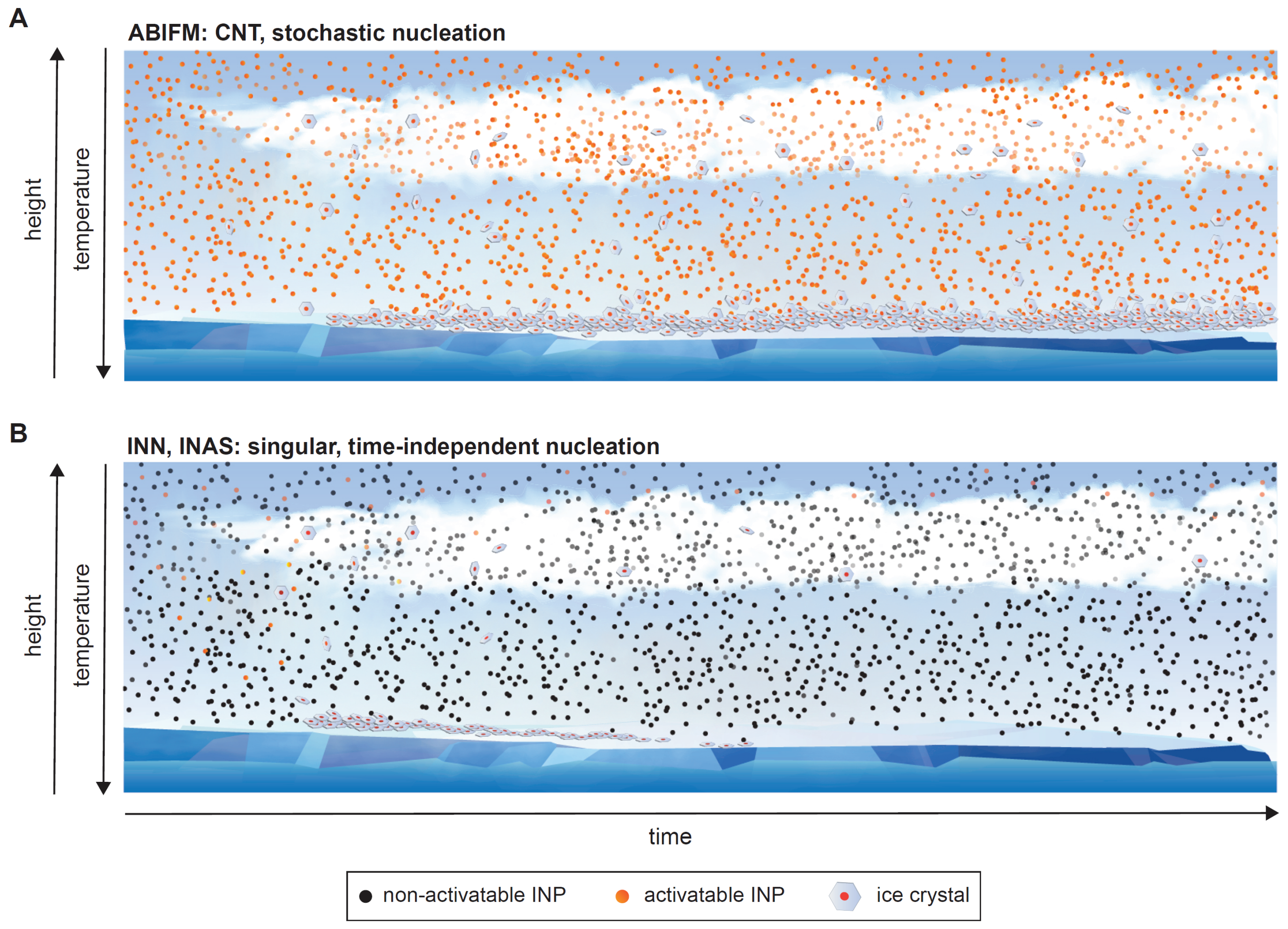
Both model environments apply time-independent (singular) and time-dependent (classical nucleation theory, CNT) parameterization schemes of immersion freezing. In the 1D model we apply three different aerosol particle types including mineral dust, sea spray aerosol (SSA), and organic (humic-like substances).

The effect of varying particle number concentration and cloud microphysical parameters, such as cloud top radiative cooling rate (CTRC), cloud top entrainment rate, and ice crystal fall velocity on the INP reservoir and ice crystal number concentrations are assessed.

We evaluate 3 commonly applied immersion freezing parameterizations which inlcude singular number-based (INN) and surface area based (INAS, ice nucleation active sites), and CNT water activity based immersion freezing model (ABIFM).

A singular approach implies instantaneously freezing (no time dependence) and unique INPs.
A CNT approach implies freezing occurs randomly among the same particles and it is time dependent, i.e., longer time at same supersaturation will yield more activated INPs.

## 1D AC Model Conceptual Approach



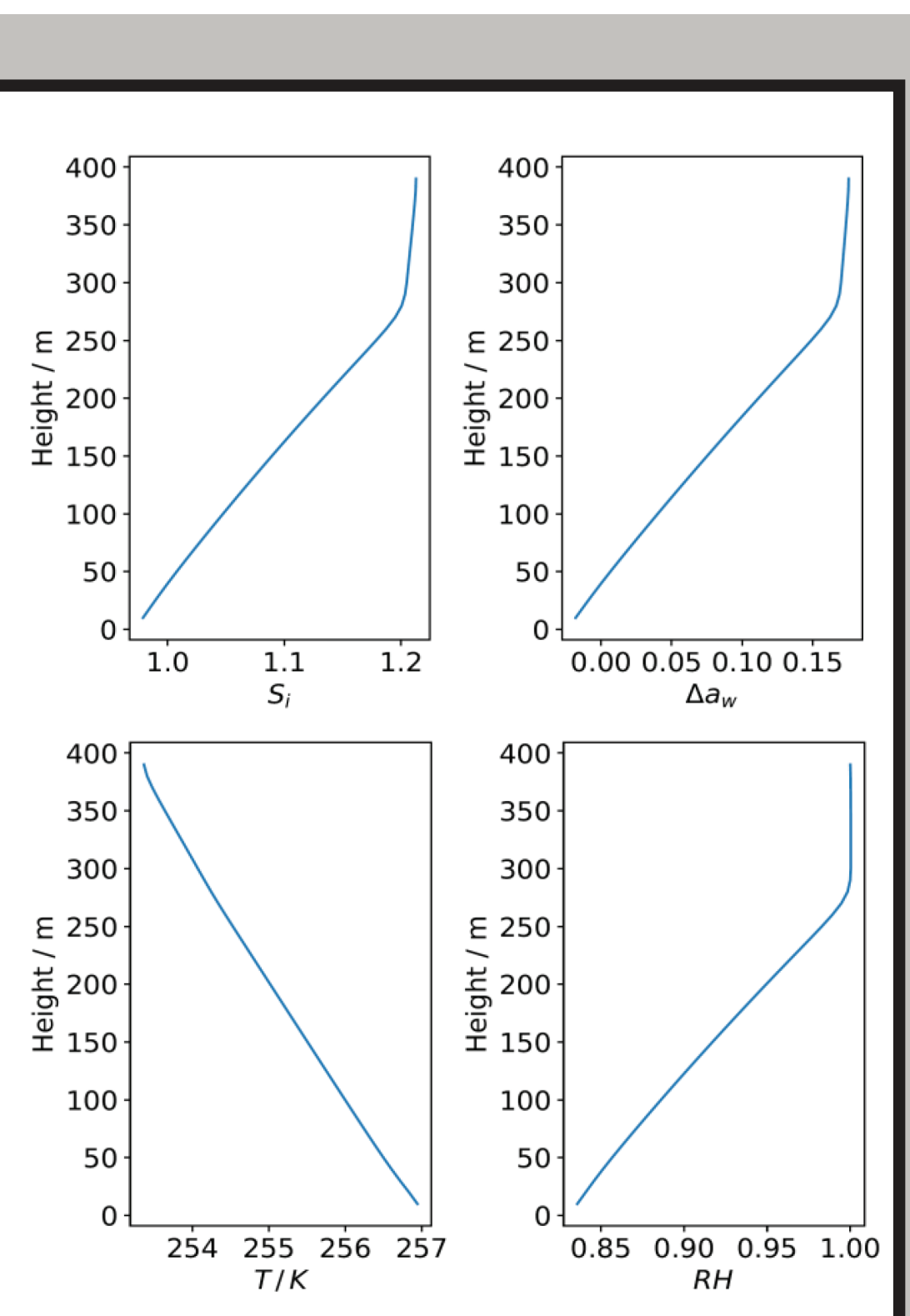
**A:** CNT-ABIFM considers stochastic, time-dependent ice nucleation. All aerosol particles are considered activatable INPs (orange circles) manifesting a large INP reservoir ($N_{INP}$). As activatable INPs enter the cloud layer, they are engulfed by supercooled water and some INPs form ICs ($N_I$). IC sedimentation reflects the loss process of the INPs.
Since INP reservoir is large and its depletion by INP activation is negligible, continuous IC formation is ensured throughout the cloud lifetime.

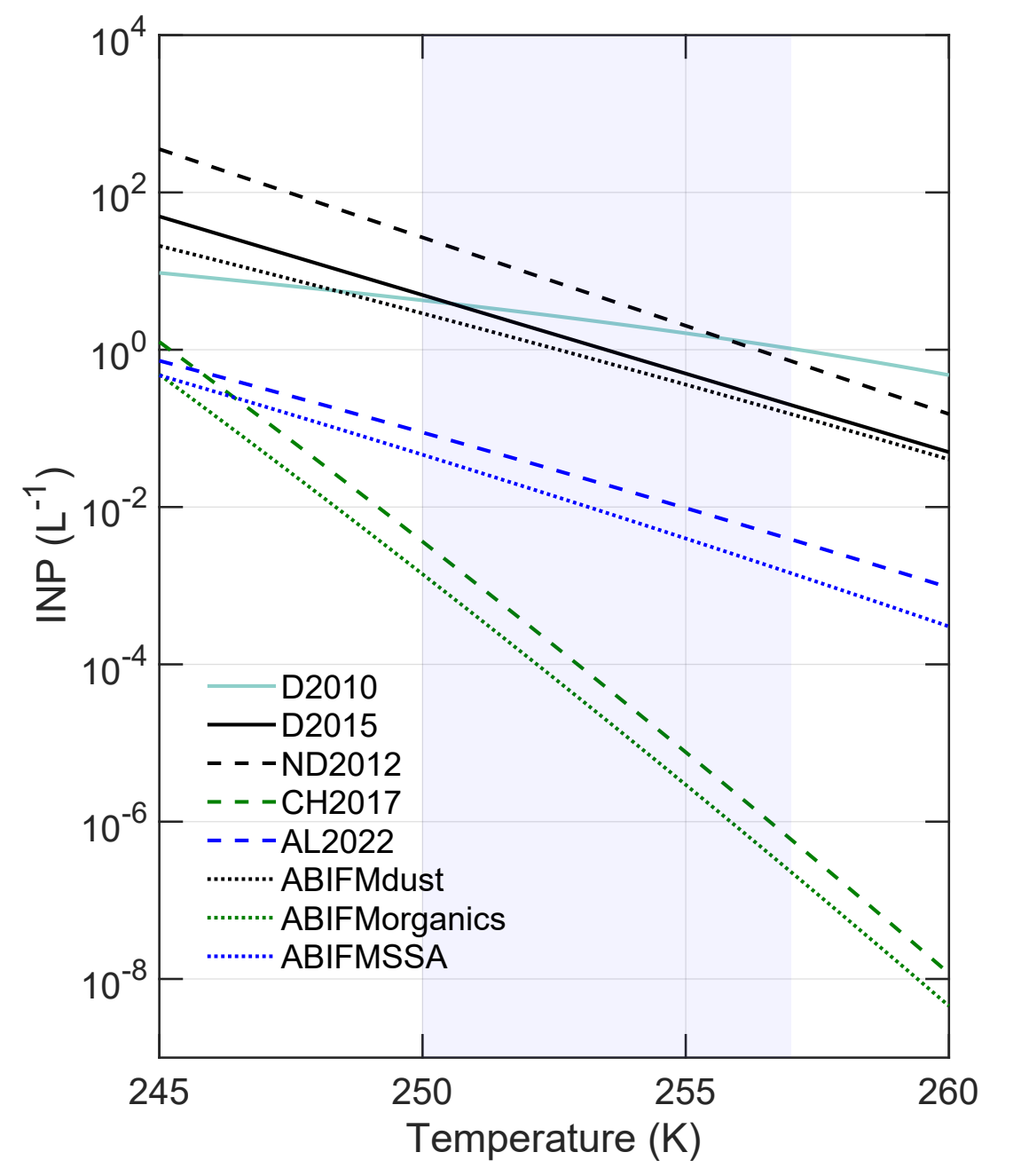
**B:** Singular INN or INAS immersion freezing parameterizations are non-stochastic and instantaneous. For given minimum cloud temperatures, only few activatable INPs out of all particles are available (few orange particles in black particle poplutation).
The few activatable INPs are quickly consumed by IC formation due to the small INP reservoir present and IC formation cannot be sustained over typical cloud lifetimes.

*Knopf et al., JAMES, in revision*

## Model Setup & INP Parameterization

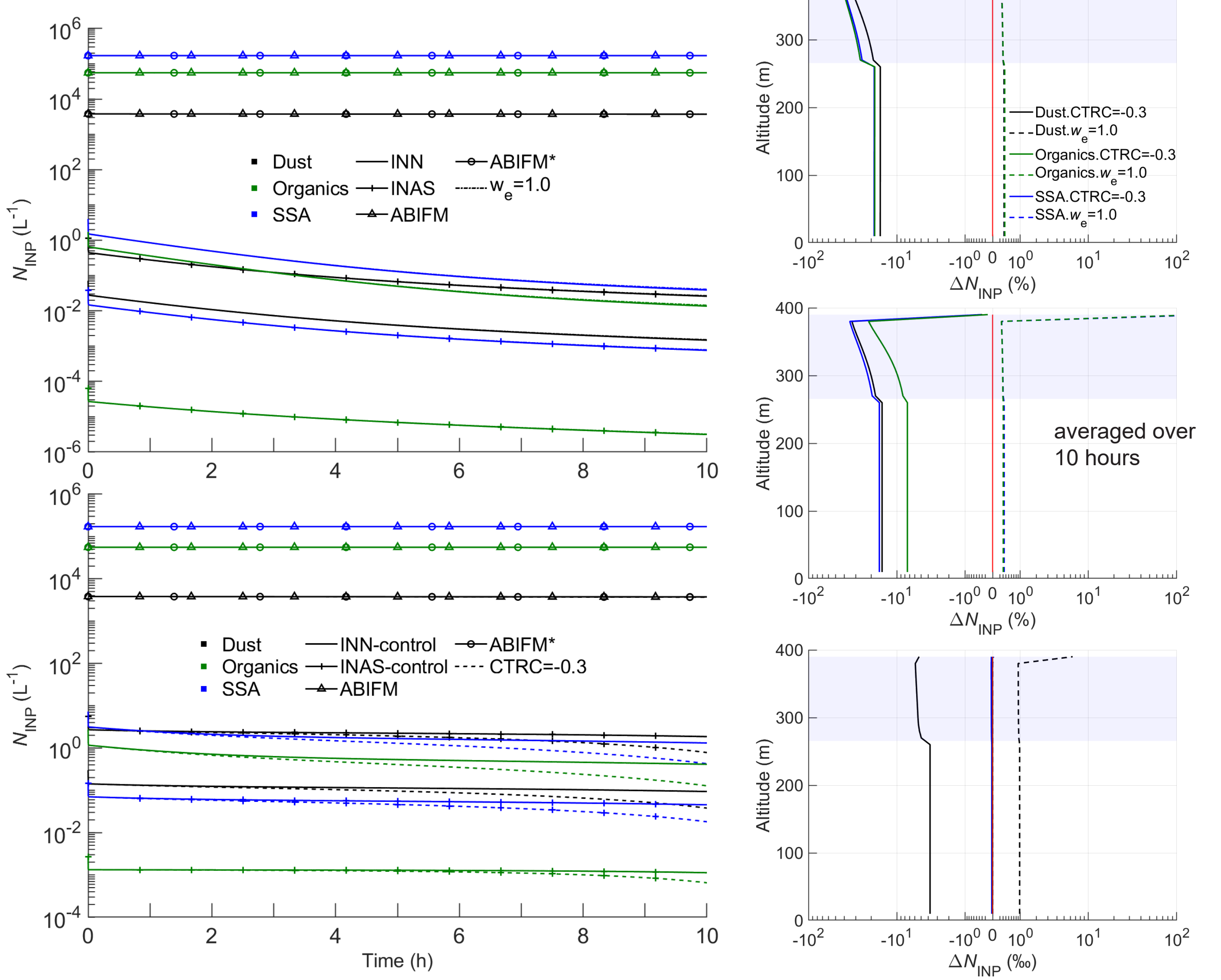
The PBL INP reservoir analysis presented builds on the Surface Heat Budget in the Arctic (SHEBA) campaign case (Fridlind et al., 2012).
The model setup includes prescribed thermodynamic profiles and represents INPs as multicomponent and polydisperse particle size distributions.
*Applied standard conditions* (which are varied)*:*
Mixing time scale: 1800 s  Entrainment rate: 0.1 cm $s^{-1}$
Sedimentation rate: 0.3 m $s^{-1}$



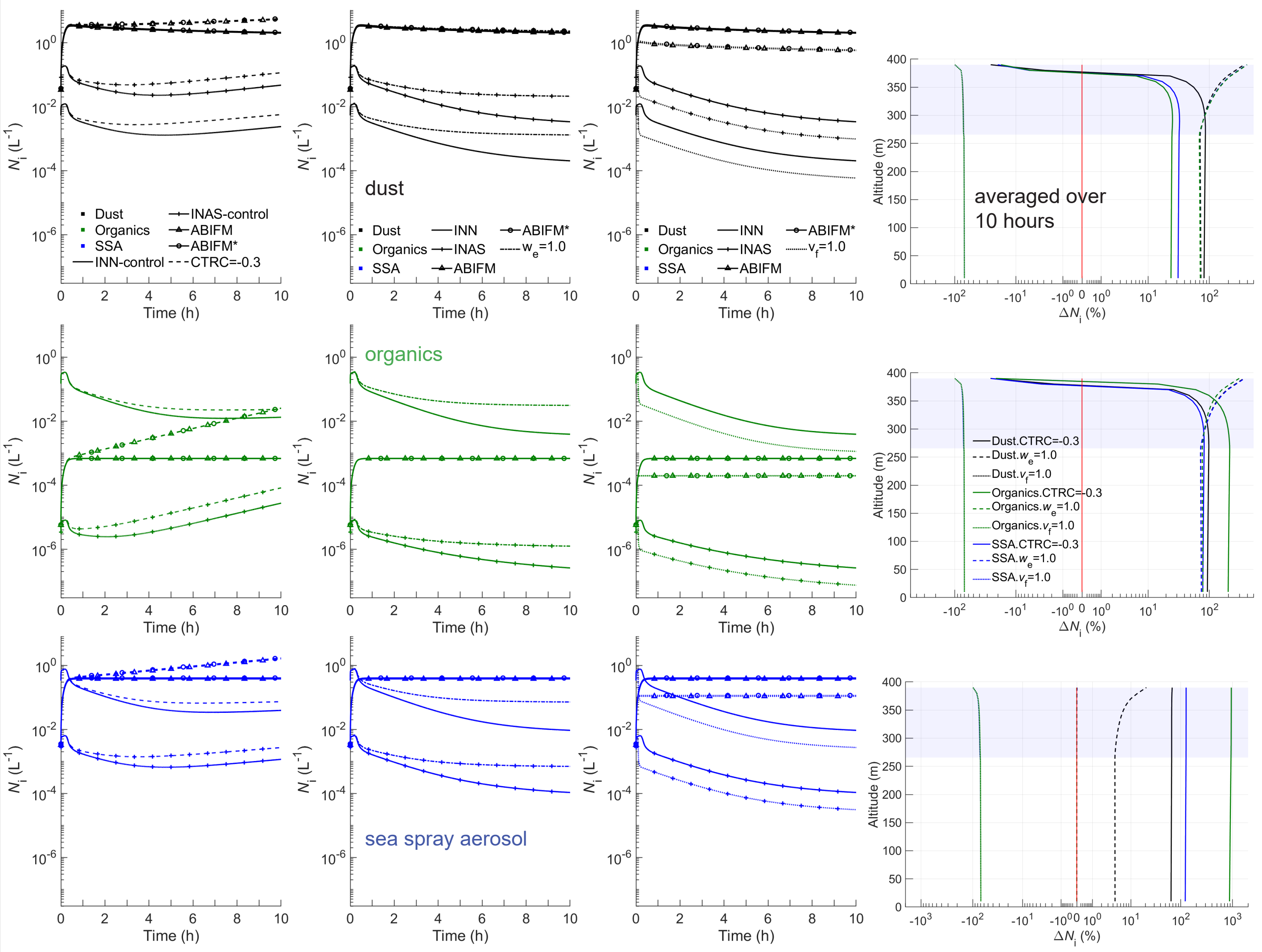


**Immersion freezing parameterizations**
*D2010*: DeMott et al. (2010), INN ambient
*D2015*: DeMott et al. (2015), INN mineral dust
*ND2012*: Niemand et al. (2012), INAS mineral dust
*CH2017*: China et al. (2017), INAS organics
*AL2022*: Alpert et al. (2022), INAS SSA
*ABIFMdust*: Alpert and Knopf (2016), ABIFM, CNT
*ABIFMorganics*: Knopf and Alpert (2013), ABIFM, CNT
*ABIFMSSA*: Alpert et al. (2022), ABIFM, CNT

## Domain Averaged and Vertically Resolved Activatable INP Number Concentrations



## Domain Averaged and Vertically Resolved IC Number Concentrations

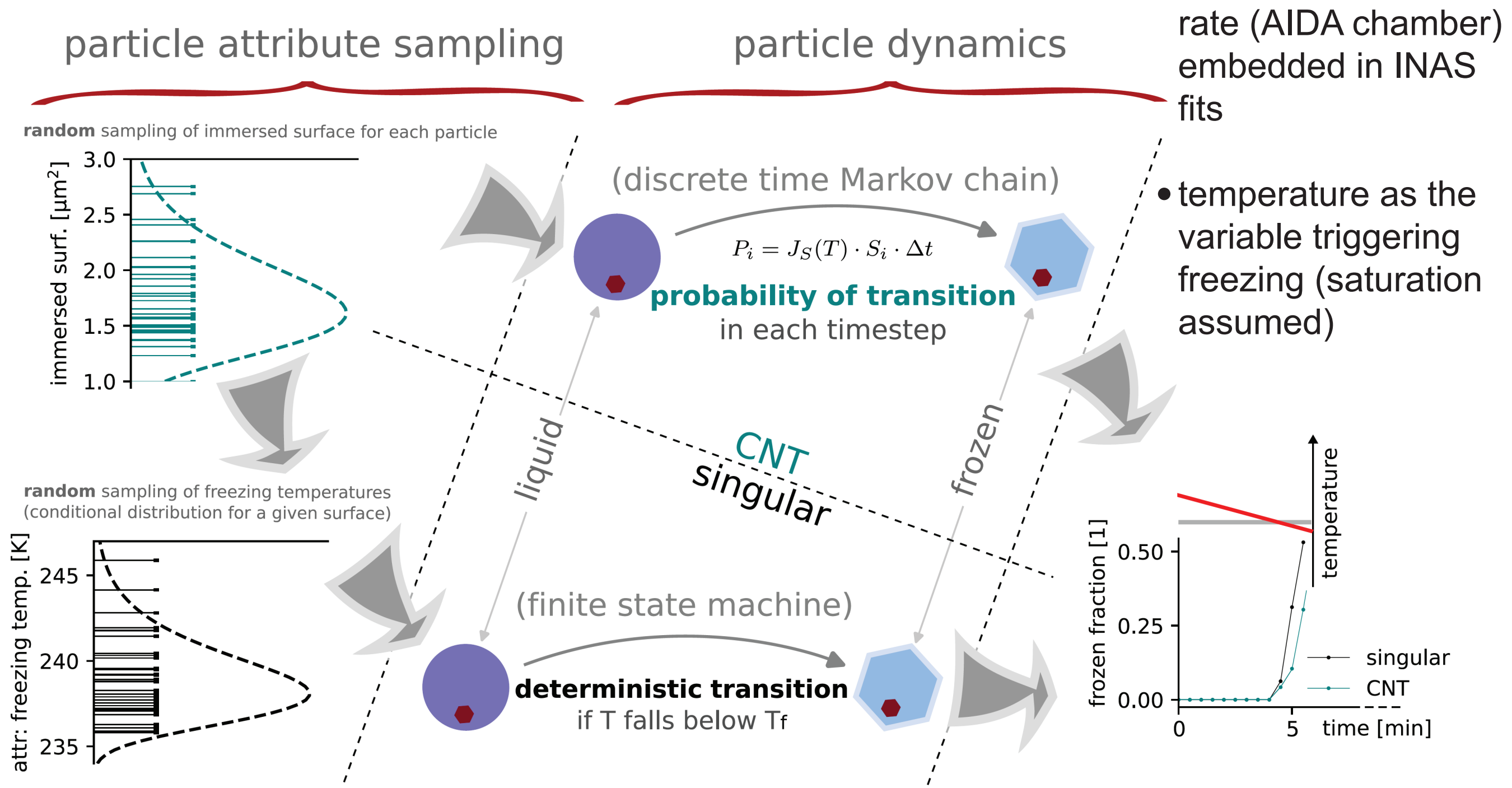


## Super-Particle Model: Concept

Aerosol particles, water droplets, and ice crystals are modelled with super-particles, each representing a large multiplicity of real world particles.

Immersion freezing is implemented using Monte-Carlo schemes:
- singular (INAS) as in Shima et al. (2020)
- CNT time dependent (ABIFM) as in Knopf and Alpert (2013)

| **singular approach** | **CNT, time-dependent approach** |
|---|---|
| • freezing temperature as particle attribute, randomly sampled at t = 0 from INAS -based on probability density function | • immersed insoluble surface as particle attribute, used to evaluate freezing probability in each step |
| • instrument cooling rate (AIDA chamber) embedded in INAS fits | • actual cooling rate in the simulated environment used |
| • temperature as the variable triggering freezing (saturation assumed) | • freezing can be triggered by employing dependence on supersaturation |



## Super-Particle Model - Nucleation Theory

*Levine (1950)*: each singularity, "mote", causes freezing
⟶ singular hypothesis

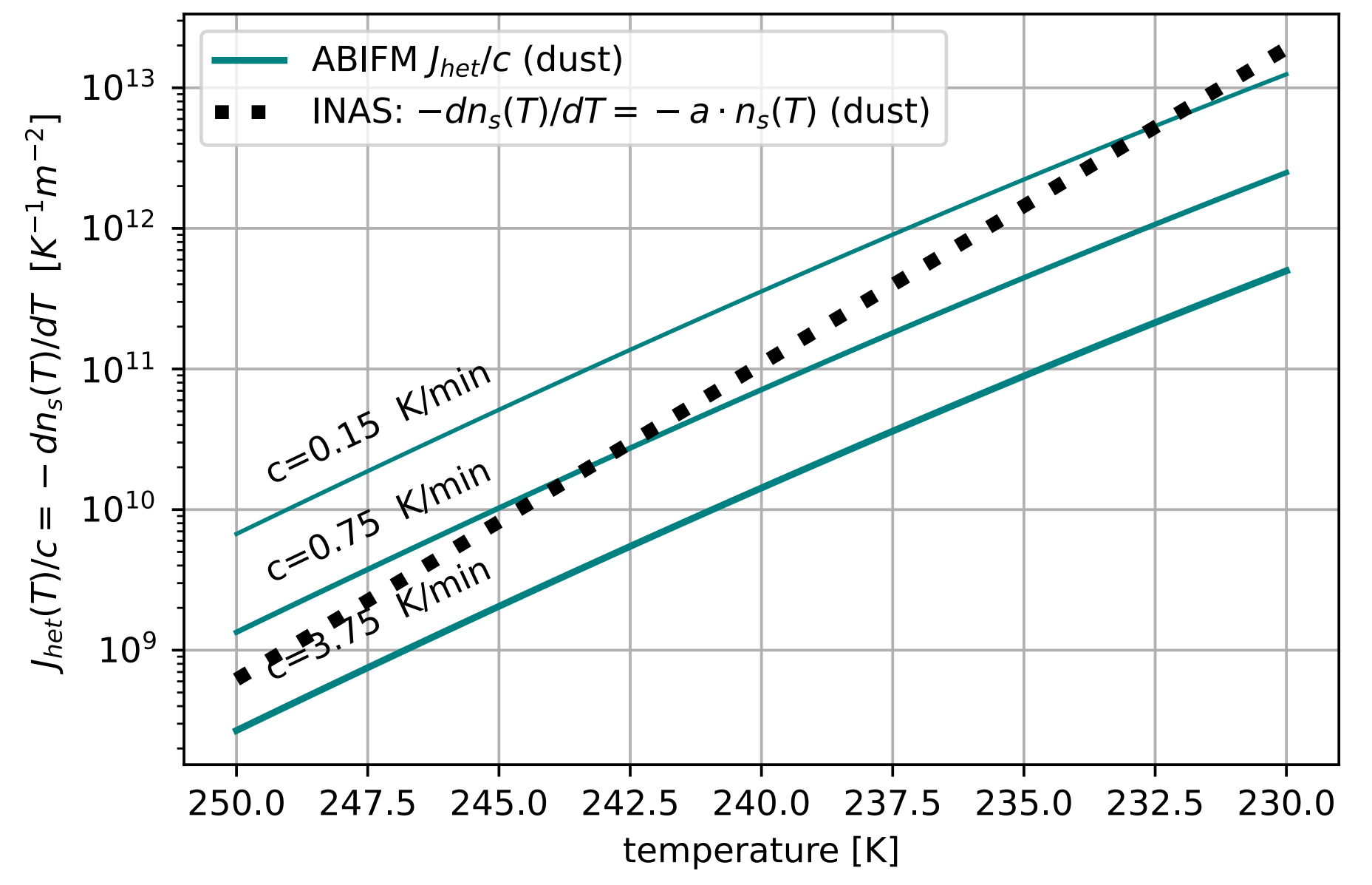
Put forward *by Biggs (1953)* and *Carte (1959)*:

$$\ln(1-P(S,t)) = -S\underbrace{\int_0^t J_{het}(T(t'))dt'}$$

Cooling rate information is lost when using INAS: $n_S(T)$

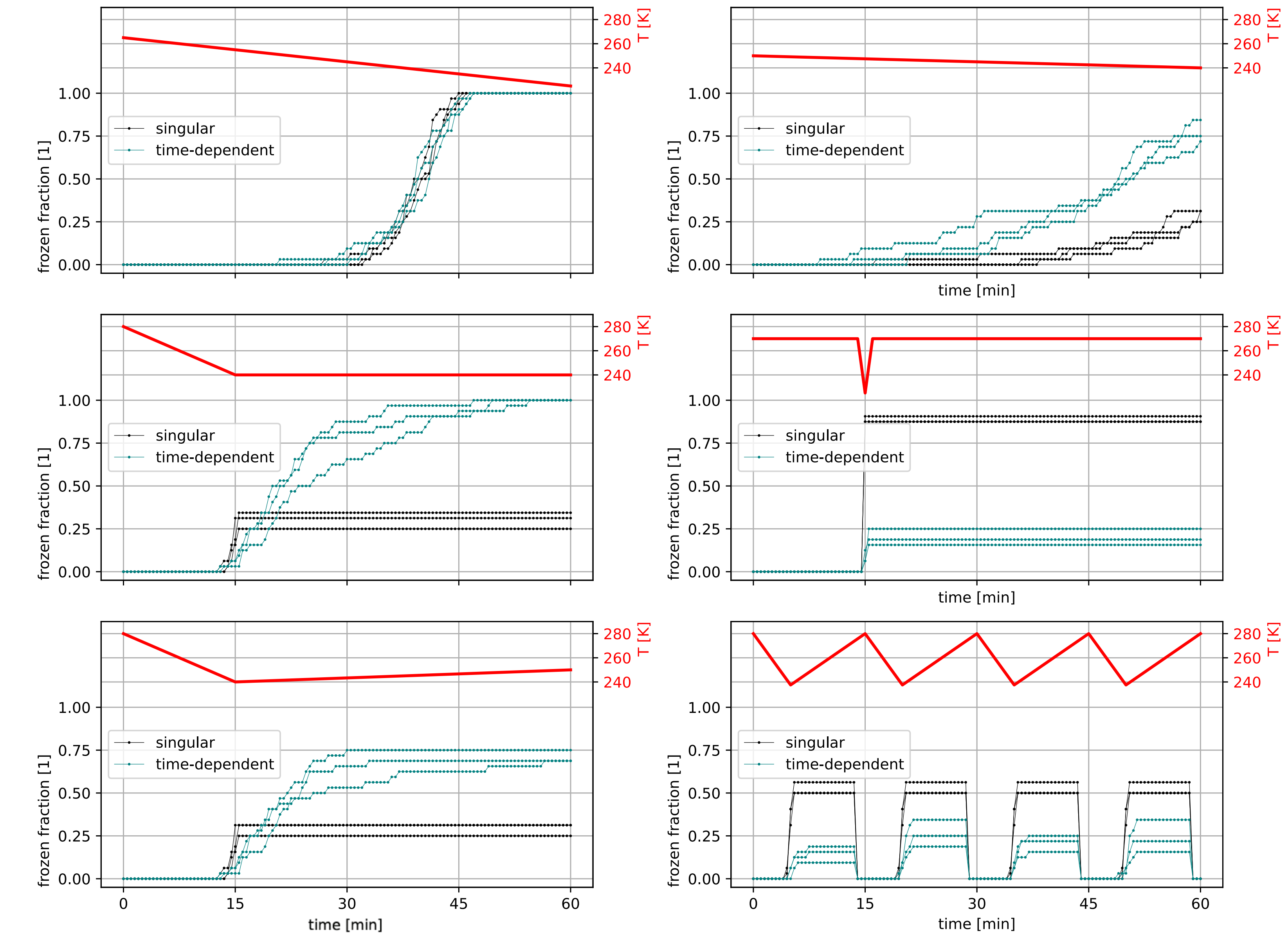
*Stansbury (1961)* and *Marshall (1961)*:
Heterogeneous nucleation is a stochastic process!

**This debate is still going on!**



INAS and CNT (ABIFM) yield same INP numbers only for experimentally accessed cooling rate. For all other conditions (as in clouds), different results will be obtained (Knopf et al., 2021).

## Super-Particle Monte-Carlo Runs: Singular vs. Time-Dependent Schemes Under Diverse Cooling Forcings - Sensitivity wrt Temperature Evolution



## Summary and Conclusions

**Minimalistic 1D Aerosol-Cloud Model (***Knopf et al., JAMES, in revision)*

- We applied a minimalistic 1D aerosol-cloud (1D AC) model to evaluate the INP reservoir dynamics using a prognostic INP treatment. We assessed how different immersion freezing parameterizations impact the available INP number concentrations that ultimately define the IC number concentrations while changing aerosol types and cloud parameters.
- The particle type and associated PSD have the greatest impact on the INP reservoir, IC number concentrations, and IC formation rates.
- INP reservoir, IC number concentrations, and IC formation rates show greatest sensitivity to cloud top radiative cooling.
- In the case of CNT (ABIFM), the IC budget is mostly affected by cloud top radiative cooling and IC sedimentation rate.
- CNT-based description results in many orders of magnitude greater INP reservoir, and sustains more than an order of magnitude greater IC number concentrations over 10-h lifetimes.

**Super-Particle Model** *(Arabas et al., in prep.)*

- A probabilistic particle-based cloud microphysics model for immersion freezing allows to examine the differences in singular and CNT-based approaches on INP and IC number concentrations.
- The 70 years ongoing debate of cooling rate impact on IC number concentration has been simulated considering different temperature profiles consistent with cloud chamber experiments and cloud conditions.
- Singular and CNT-based approaches produce the same number of IC only for the cooling rates that were applied when deriving those parameterizations. Otherwise, large difference in IC number concentrations are observed.

**General**

- The choice of applied immersion freezing parameterization impacts model results significantly, and, thus, should be a matter of further research.
- Prognostic treatment of INPs in the 1D AC model is computationally less costly when applying a CNT approach, whereas the opposite is the case for the super-particle model. However, in both applications the CNT approach provides a more robust description when different cloud microphysical time scales are considered.

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