Immersion freezing in particle-based cloud μ-physics models

S. Arabas[1] (ex-2), J.H. Curtis[2], I. Silber[3], A. Fridlind[4], D.A. Knopf[5], M. West[2] & N. Riemer[2]

1: agh.edu.pl
2: atmos.illinois.edu
3: pnnl.gov
4: giss.nasa.gov
5: stonybrook.edu

funding:
ASR Atmospheric System Research
National Science Centre Poland
Excellence Initiative Research University

University of Illinois, Urbana-Champaign, Aug 26th 2025

JAMES | Journal of Advances in Modeling Earth Systems

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Immersion Freezing in Particle-Based Aerosol-Cloud Microphysics: A Probabilistic Perspective on Singular and Time-Dependent Models

Sylwester Arabas[1], Jeffrey H. Curtis[2], Israel Silber[3,4], Ann M. Fridlind[5], Daniel A. Knopf[6], Matthew West[7], and Nicole Riemer[2]

10.1029/2024MS004770

Aerosol-cloud interactions: a conceptual picture

Aerosol-cloud interactions: a conceptual picture

background image: vitsly / Hokusai

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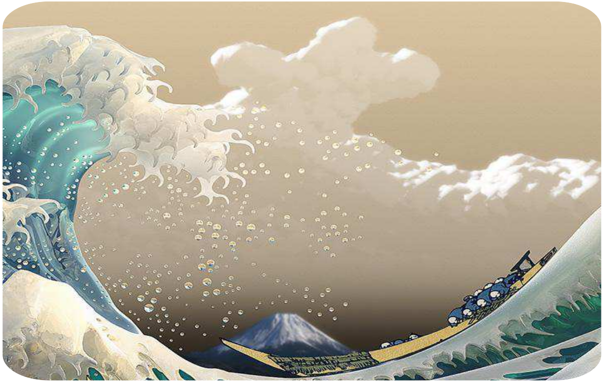

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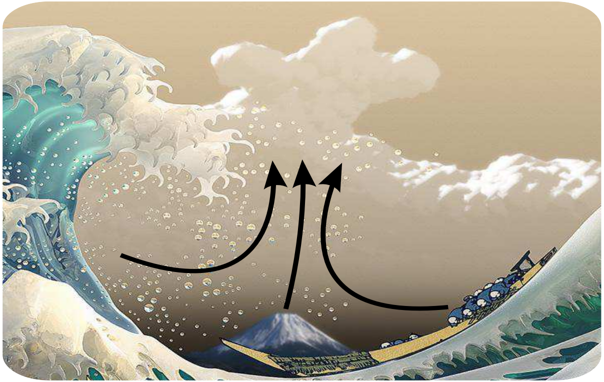
Aerosol-cloud interactions: a conceptual picture

- aerosol particles of natural
  and anthropogenic origin
  act as condensation/crystallisation nuclei



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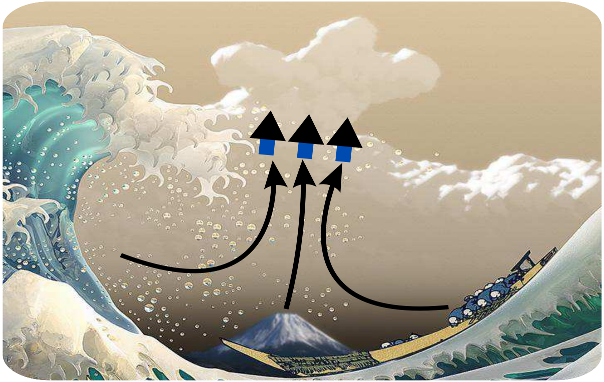

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two-way interactions:
- aerosol characteristics influence cloud microstructure
- cloud processes influence aerosol size and composition

background image: vitsly / Hokusai

Aerosol-cloud interactions: μ-physics models

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COMMISSIONED MANUSCRIPT
10.1029/2019MS001689

Key Points:
- Microphysics is an important component of weather and climate models, but its representation in current models is highly uncertain

Confronting the Challenge of Modeling Cloud and Precipitation Microphysics

Hugh Morrison[1], Marcus van Lier-Walqui[2], Ann M. Fridlind[3], Wojciech W. Grabowski[1], Jerry Y. Harrington[4], Corinna Hoose[5], Alexei Korolev[6], Matthew R. Kumjian[4], Jason A. Milbrandt[7], Hanna Pawlowska[8], Derek J. Posselt[9], Olivier P. Prat[10], Karly J. Reimel[4], Shin-Ichiro Shima[11], Bastiaan van Diedenhoven[2], and Lulin Xue[1]

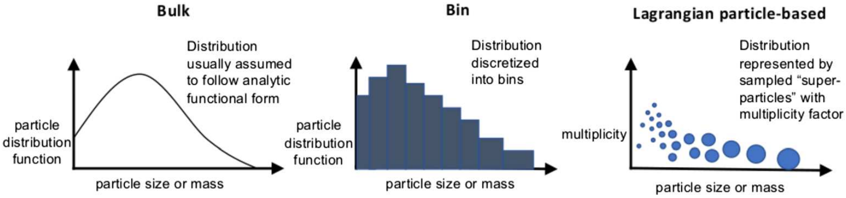
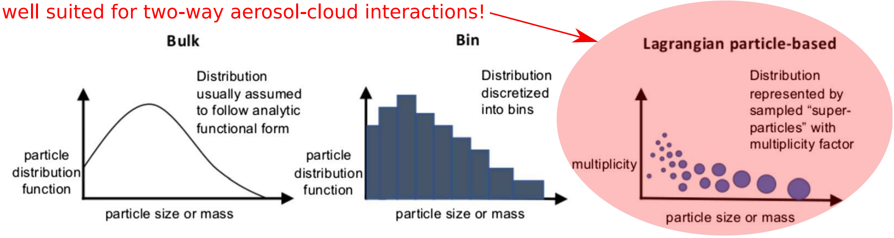


Figure 3. Representation of cloud and precipitation particle distributions in the three main types of microphysics

Aerosol-cloud interactions: μ-physics models

well suited for two-way aerosol-cloud interactions!



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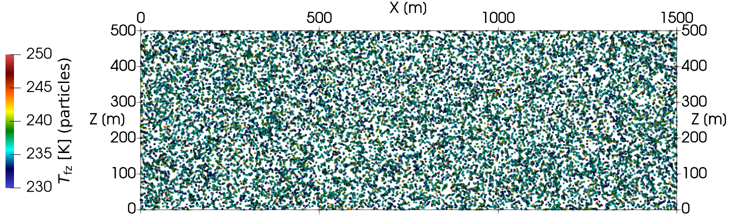
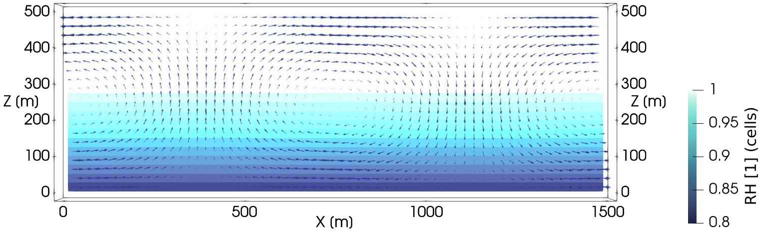
Particle-based μ-physics + prescribed-flow: model state

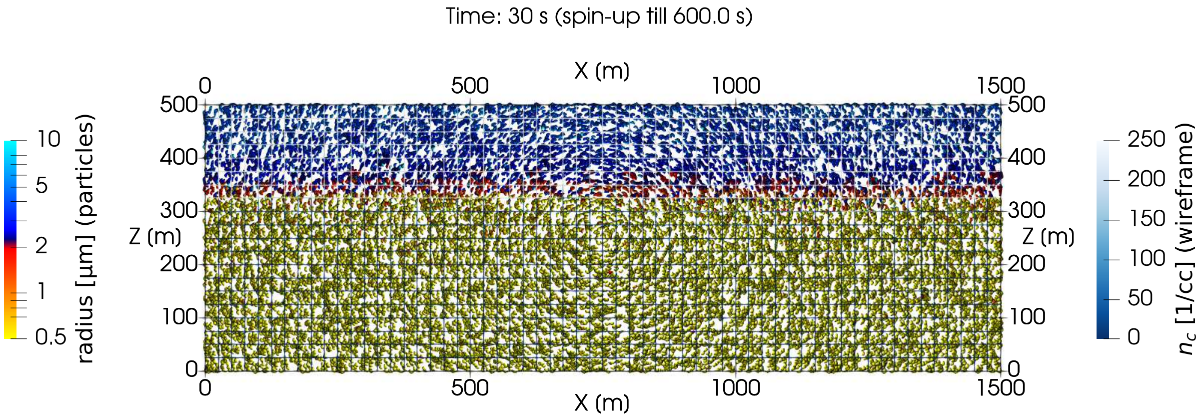
Lagrangian component



Eulerian component



Particle-based μ-physics + prescribed-flow: spin-up



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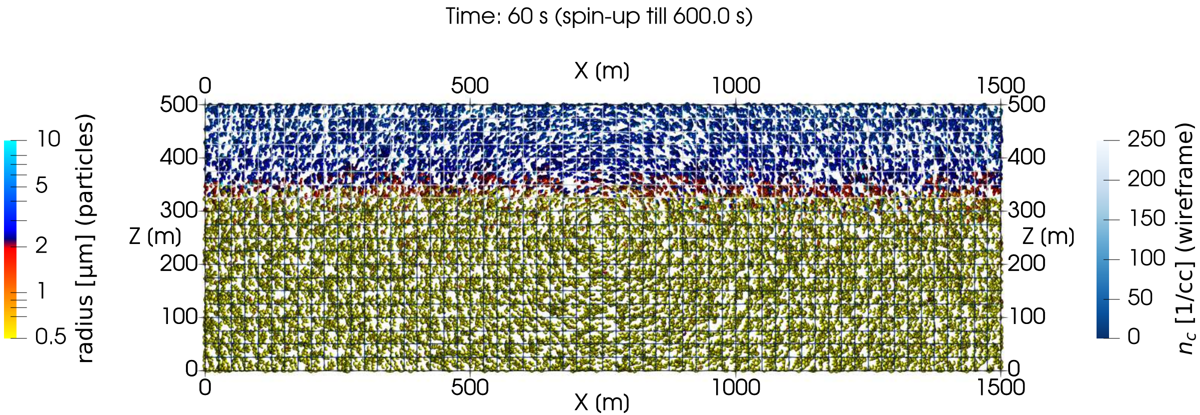

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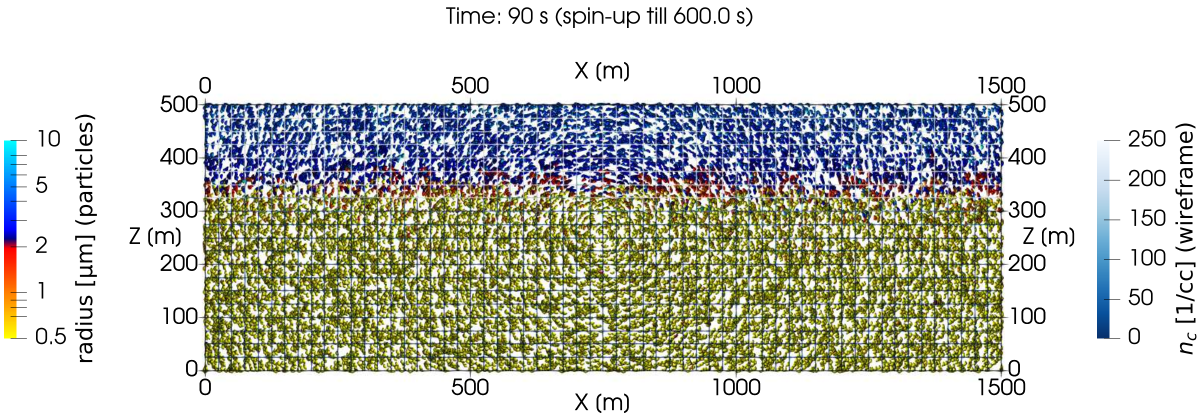

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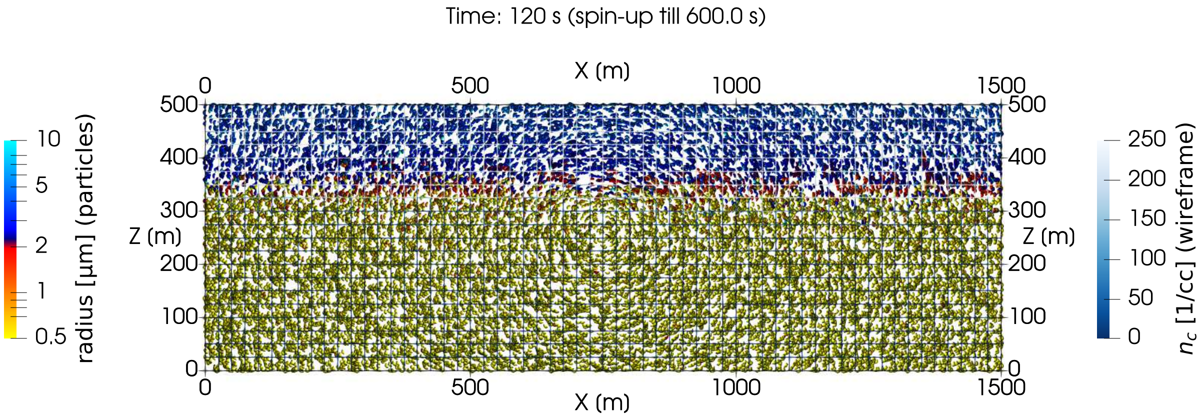

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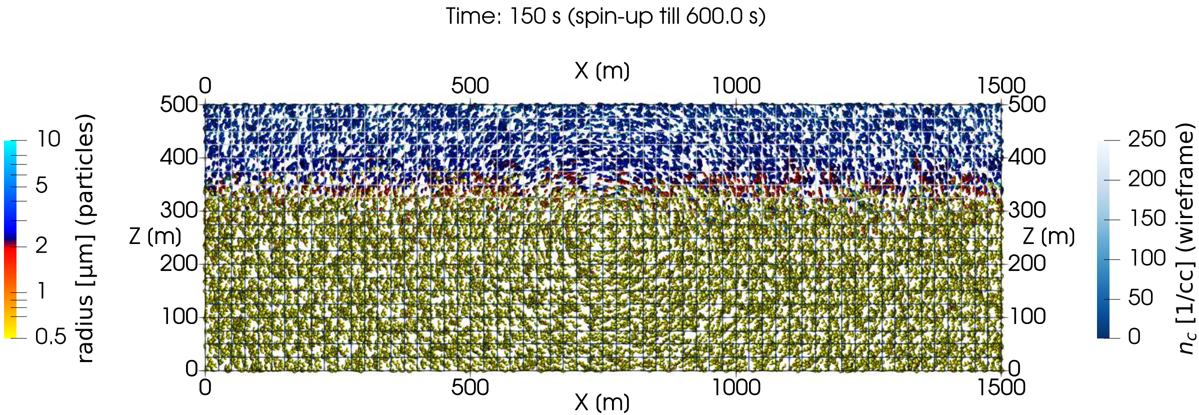

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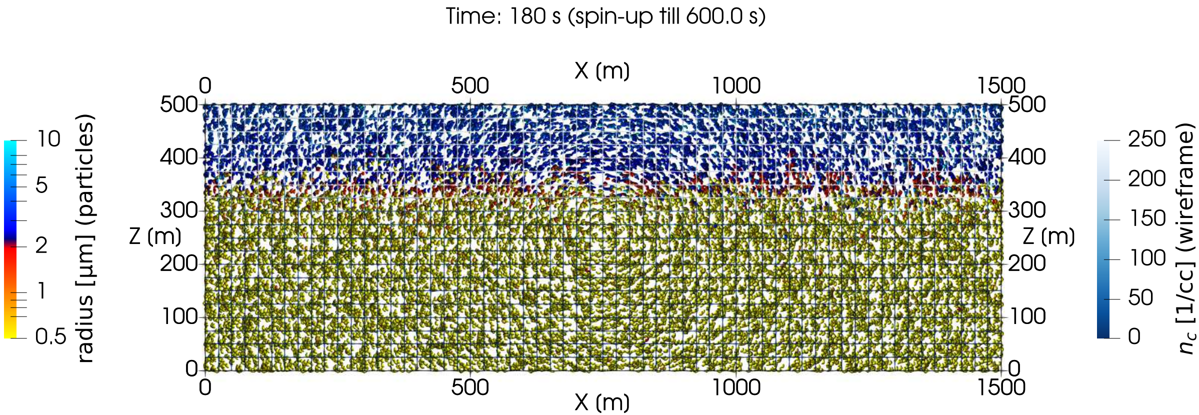

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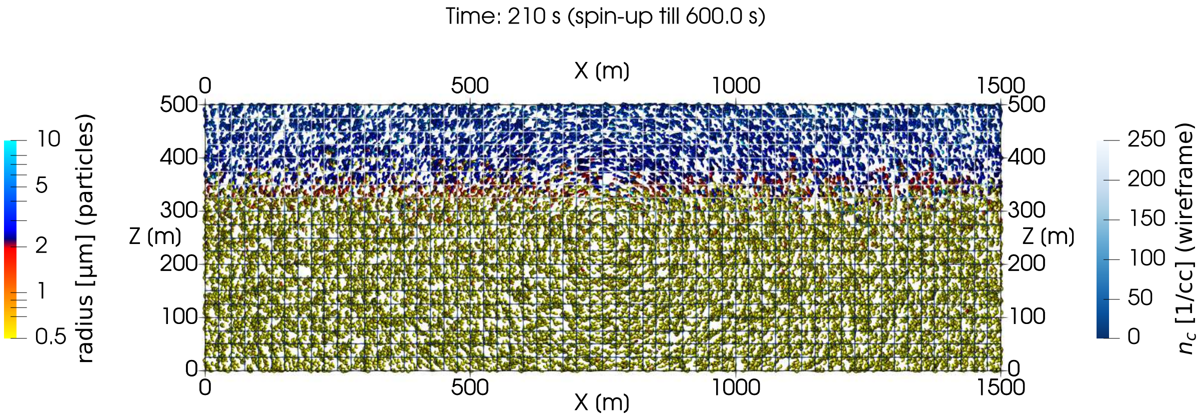

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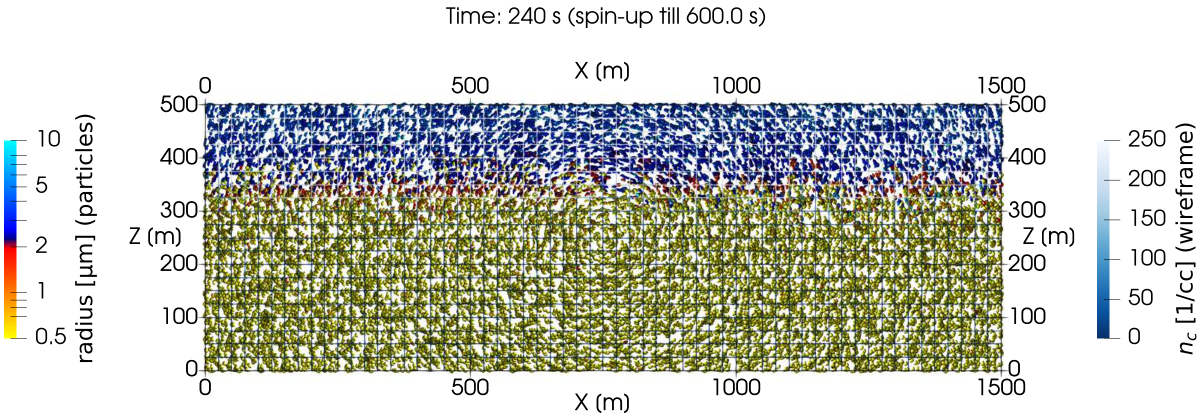

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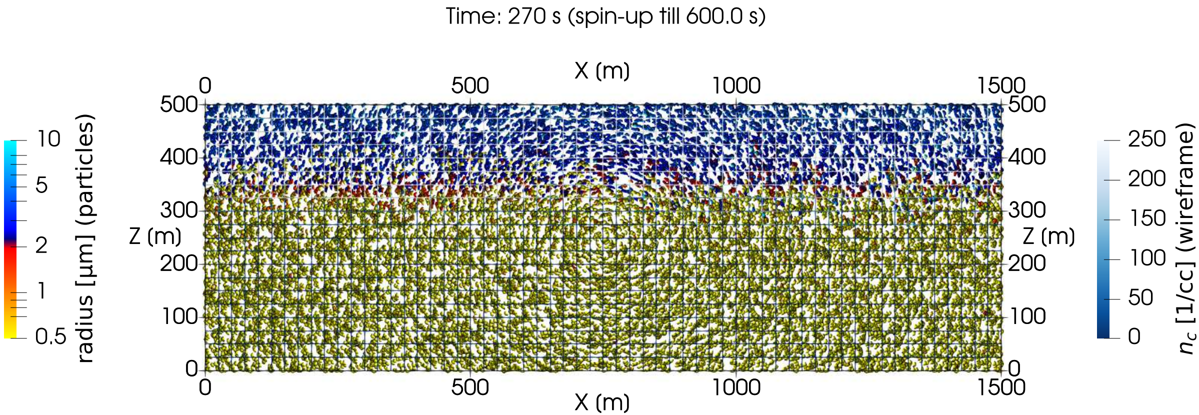

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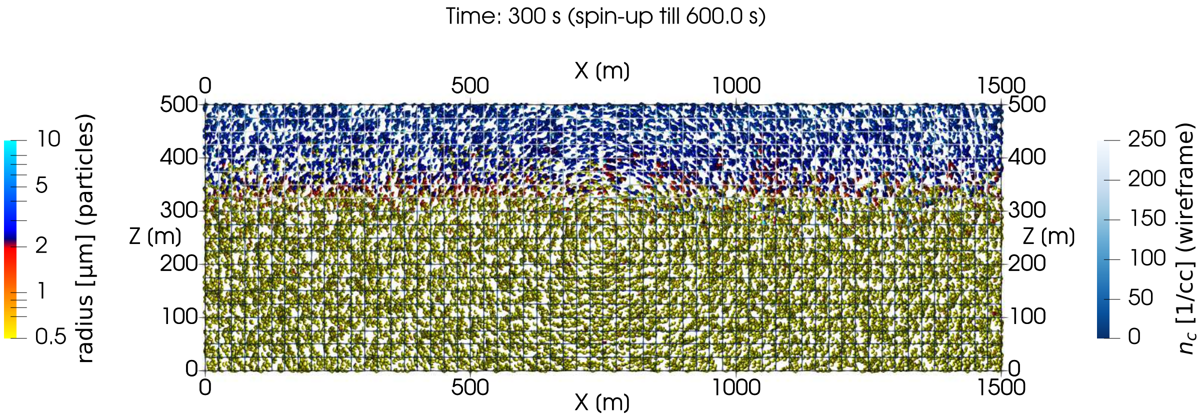
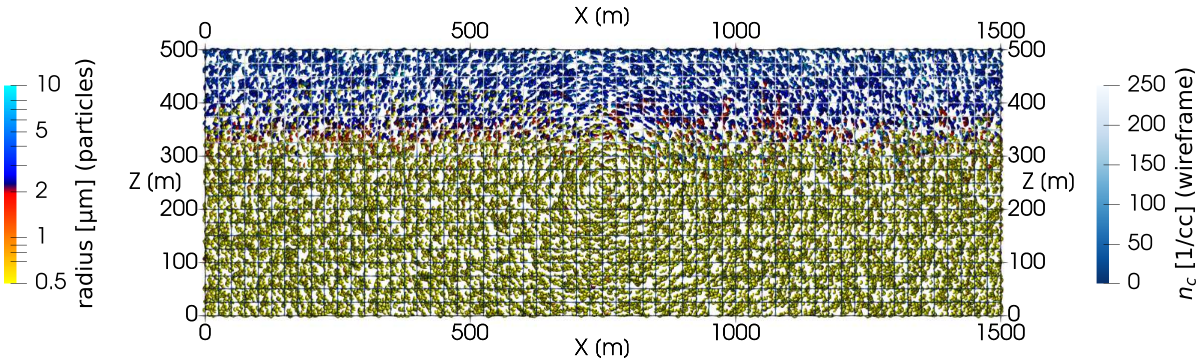


Particle-based μ-physics + prescribed-flow: spin-up

Time: 330 s (spin-up till 600.0 s)



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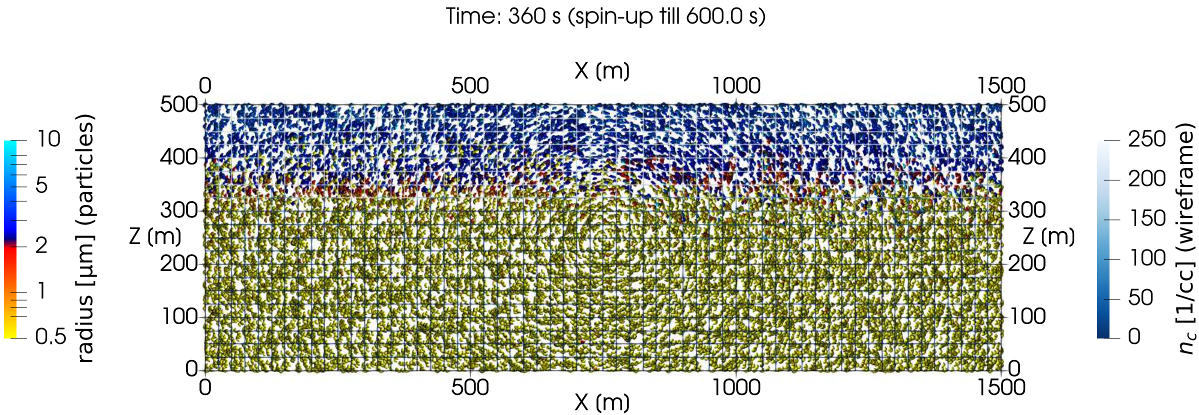
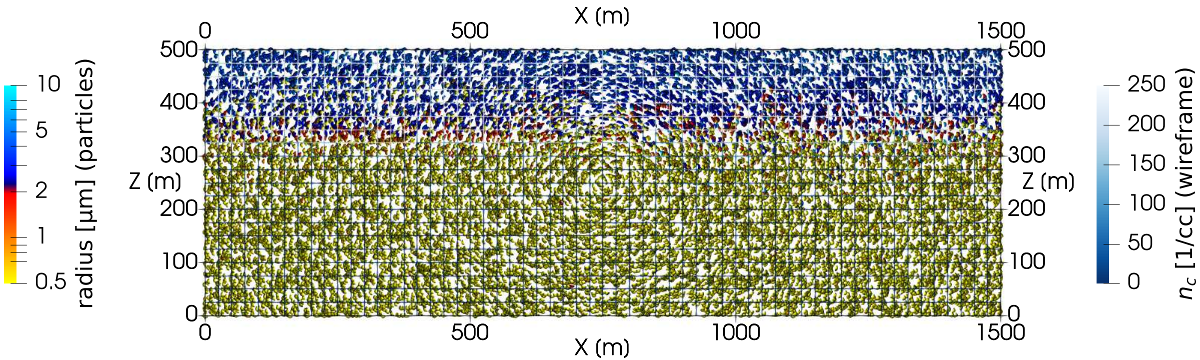


Particle-based μ-physics + prescribed-flow: spin-up

Time: 390 s (spin-up till 600.0 s)



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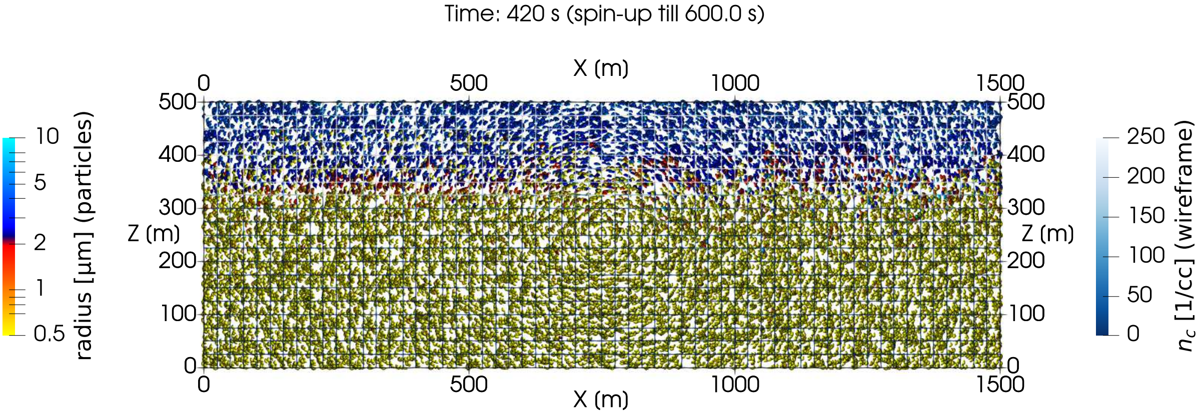

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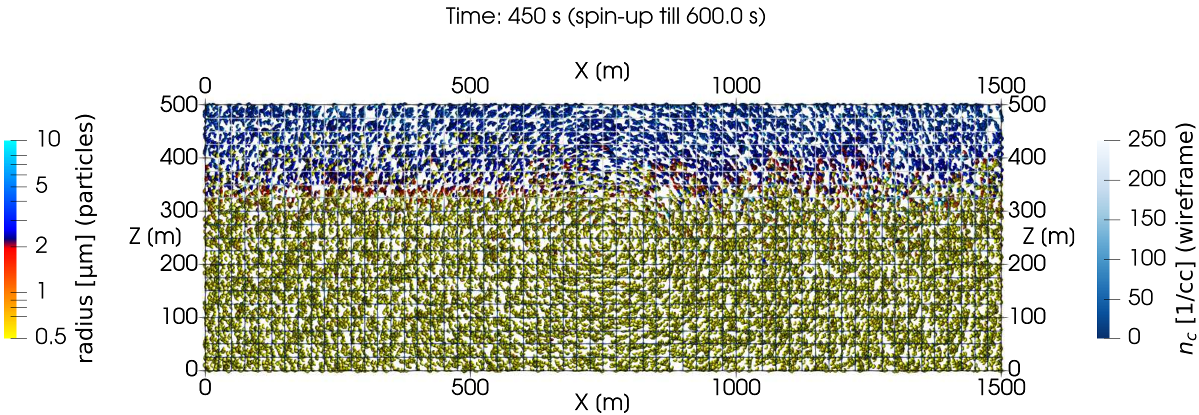

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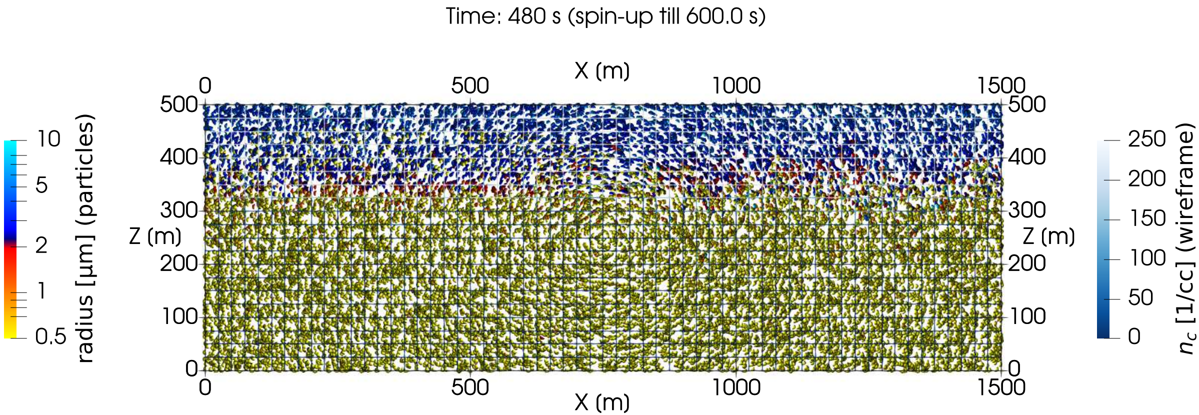

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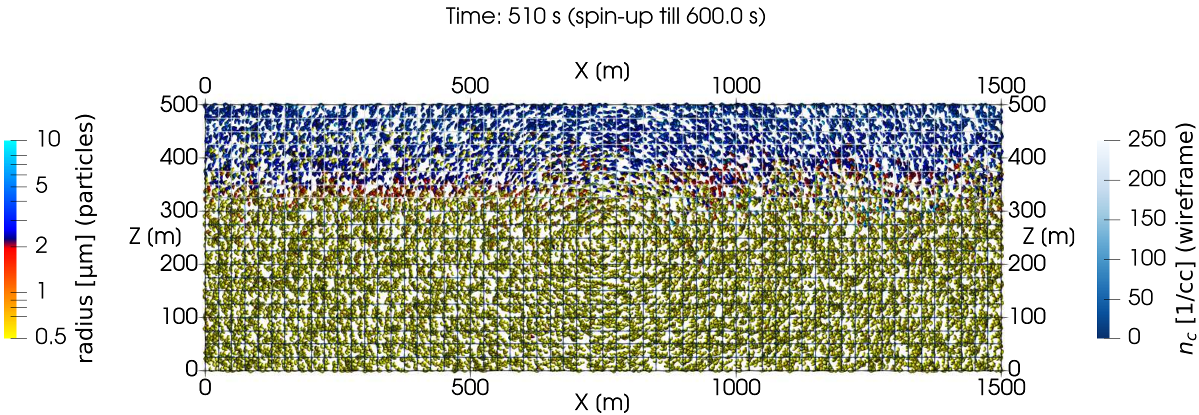

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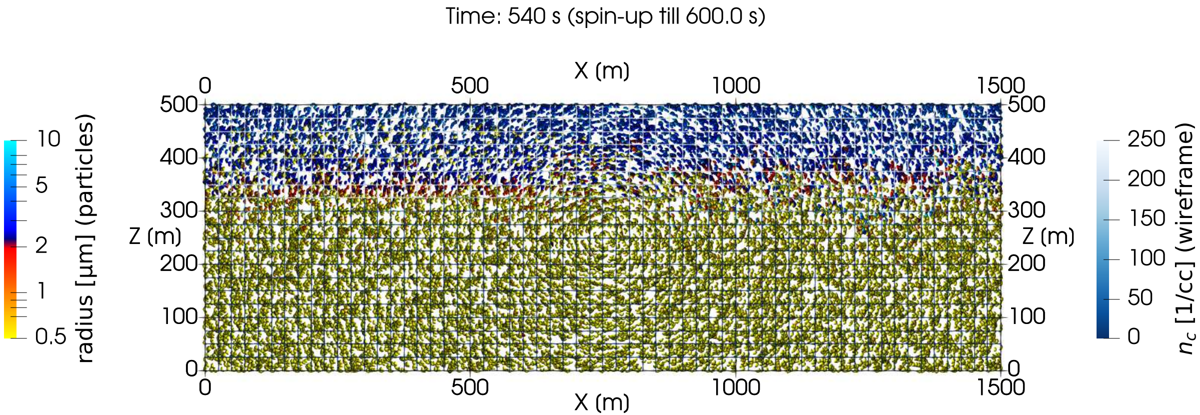
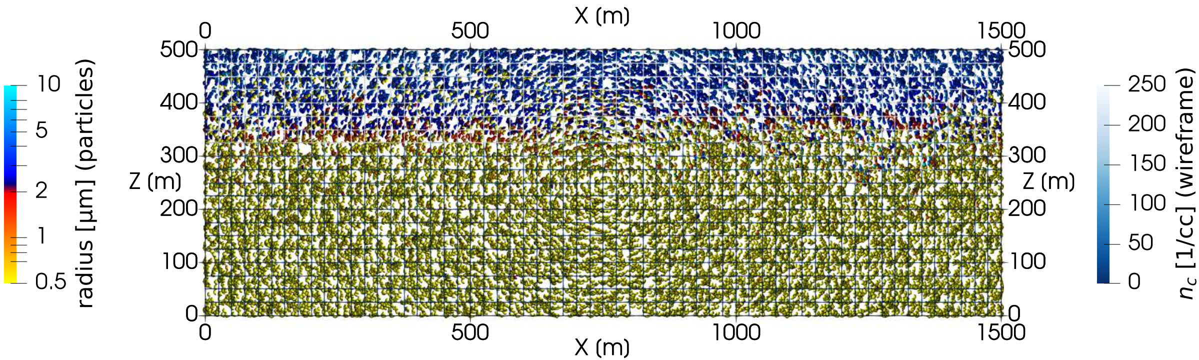


Particle-based μ-physics + prescribed-flow: spin-up

Time: 570 s (spin-up till 600.0 s)



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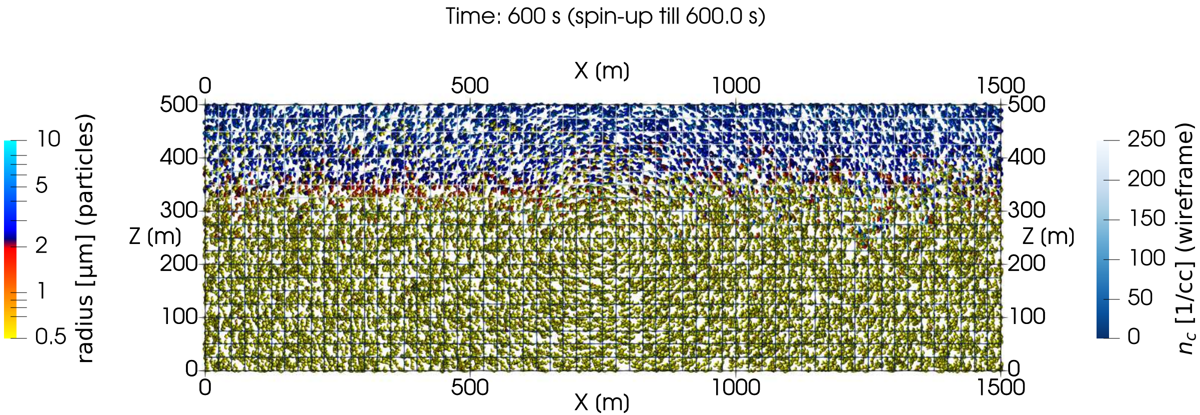

Shima et al. '20 particle-based mixed-phase μ-physics

Shima, Sato, Hashimoto & Misumi 2020 (GMD):
Predicting the morphology of ice particles in deep convection using the super-droplet method

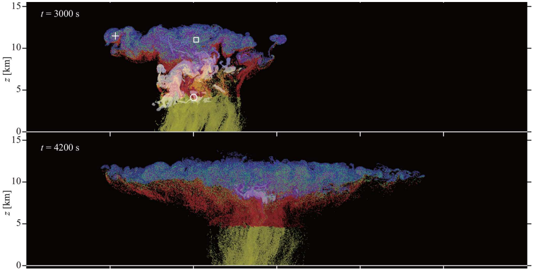

Figure 1. Typical realization of CTRL cloud spatial structures at $t = 2040$, 2460, 3000, 4200, and 5400 s. The mixing ratio of cloud water, rainwater, cloud ice, graupel, and snow aggregates are plotted in fading white, yellow, blue, red, and green, respectively. The symbols indicate examples of unrealistic predicted ice particles (Sects. 7.3 and 9.1). See also Movie 1 in the video supplement.

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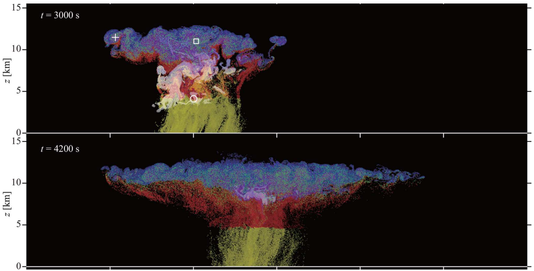

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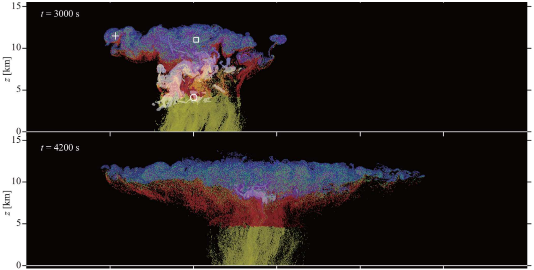

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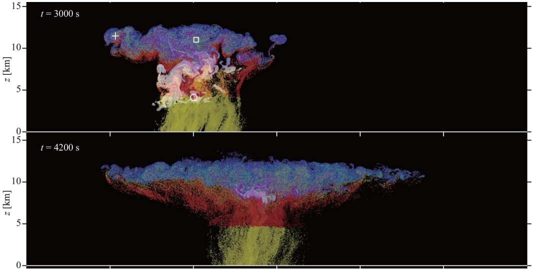

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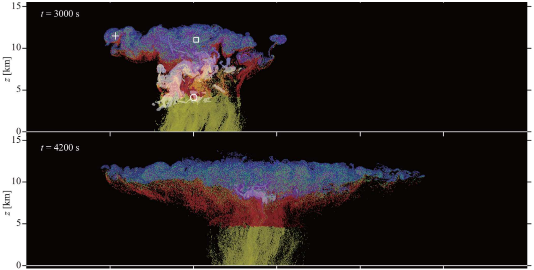

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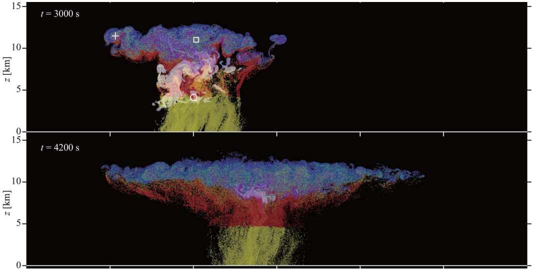

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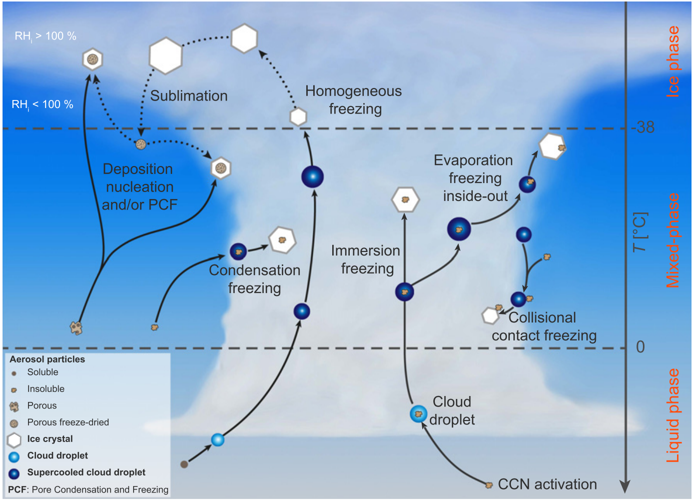
Immersion freezing and other ice crystal formation pathways in clouds



Kanji et al. 2017, graphics F. Mahrt, `https://doi.org/10.1175/AMSMONOGRAPHS-D-16-0006.1`

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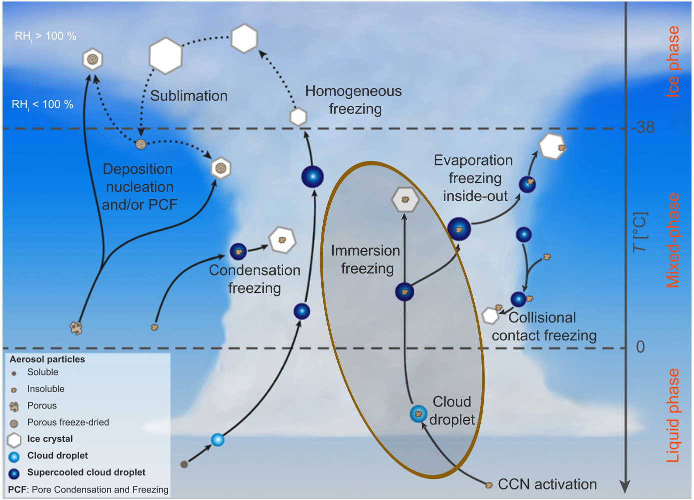

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Immersion freezing: bacteria and the Olympics

Journal of Geophysical Research: Atmospheres

RESEARCH ARTICLE

10.1002/2016JD025251

Key Points:

- Very ice active Snomax protein aggregates are fragile and their ice nucleation ability decreases over months of freezer storage
- Partitioning of ice active protein aggregates into the immersion oil reduces the droplet's measured freezing temperature

The unstable ice nucleation properties of Snomax® bacterial particles

Michael Polen[1], **Emily Lawlis**[1], **and Ryan C. Sullivan**[1]

[1]Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, Pennsylvania, USA

Abstract Snomax® is often used as a surrogate for biological ice nucleating particles (INPs) and has recently been proposed as an INP standard for evaluating ice nucleation methods. We have found the immersion freezing properties of Snomax particles to be substantially unstable, observing a loss of ice nucleation ability

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https://www.reuters.com/markets/commodities/making-snow-stick-wind-challenges-winter-games-slope-makers-2021-11-29/

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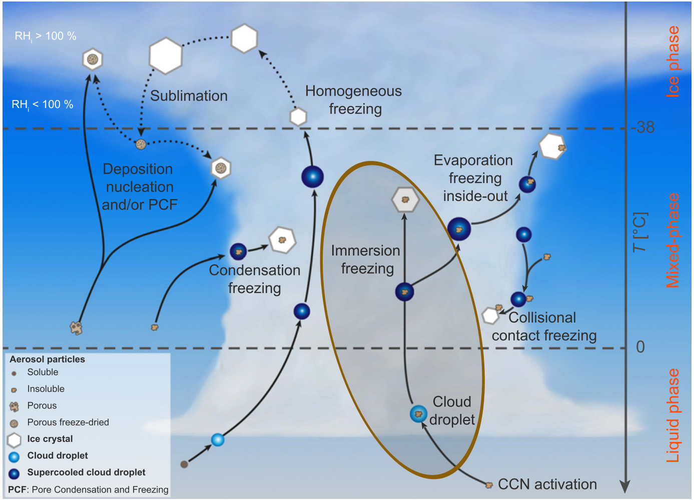

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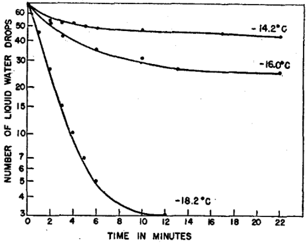
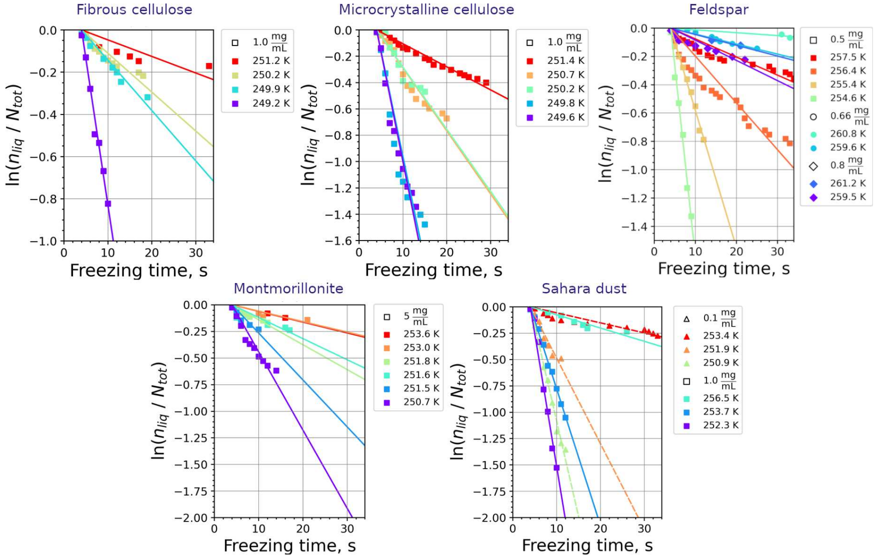
Vonnegut 1948 (J. Colloid Sci.)



Fraction of water drops remaining unfrozen as a function of time.

Szakáll et al. 2021, ACP 21: isothermal experiments



Marshall et al. 1961, Nubila 4(1)

Heterogeneous Nucleations is a Stochastic Process

by

J. S. MARSHALL

McGill University, Montreal, Canad.

Presented at the International Congress on the Physics of Clouds (Hailstorms) at Verona 9-13 August 1960.

Poissonian model of freezing & Ice Nucleation Active Sites (INAS)

theory (in modern notation)
(Bigg '53, Langham & Mason '58, Carte '59, Marshall '61)

Poisson counting process with rate r:

$$P^*(\text{k events in time t}) = \frac{(rt)^k \exp(-rt)}{k!}$$

$$P(\text{one or more events in time t}) = 1 - P^*(k=0, t)$$

$$\ln(1-P) = -rt$$

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introducing $J_{\text{het}}(T)$, $T(t)$ and INP surface A:

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

Poissonian model of freezing & Ice Nucleation Active Sites (INAS)

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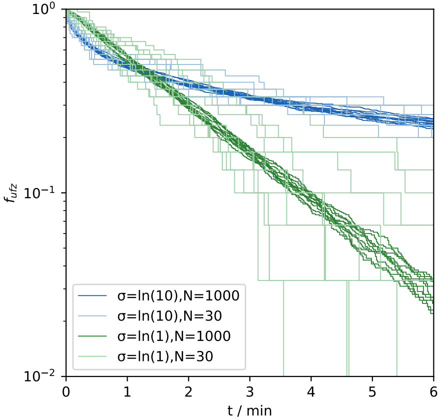
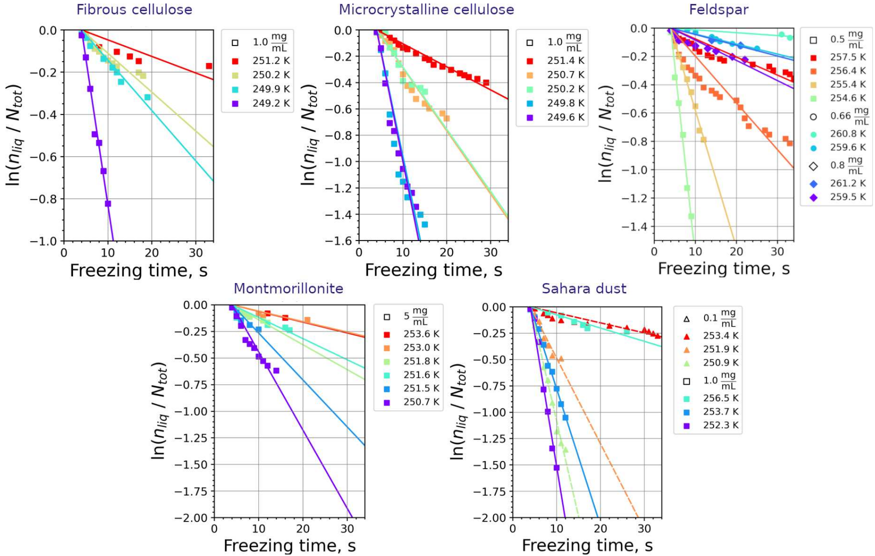


(as in Alpert & Knopf 2016, Fig. 1a)

Szakáll et al. 2021, ACP 21: isothermal experiments



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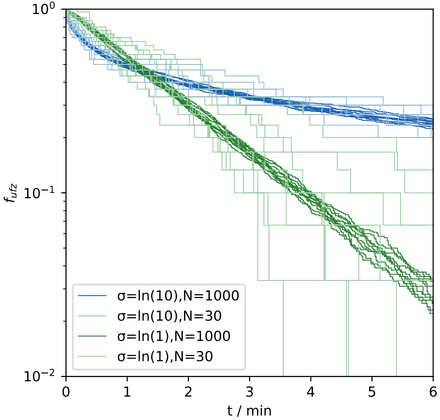

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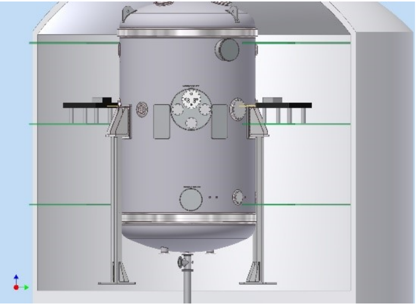
AIDA @ KIT



(https://www.imk-aaf.kit.edu/, photo: KIT/Ottmar Möhler)

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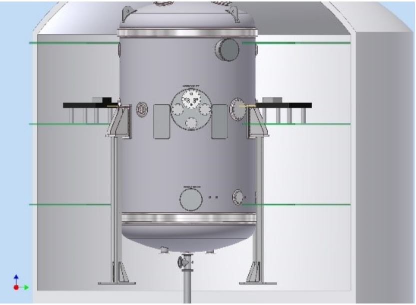

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AIDA cooling rate: ca. 0.5 K/min

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J_{het} or n_s ?

Vali 2014 (ACP)

"Interpretations of the experimental results face considerable difficulties ... two separate ways of interpreting the same observations; one assigned primacy to time the other emphasized the temperature-dependent impacts of the impurities ... dichotomy – the stochastic and singular models"

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JOURNAL OF GEOPHYSICAL RESEARCH: ATMOSPHERES, VOL. 118

Minimal cooling rate dependence of ice nuclei activity in the immersion mode

Timothy P. Wright,[1] Markus D. Petters,[1] John D. Hader,[1] Travis Morton,[1] and Amara L. Holder[1]

Received 12 April 2013; revised 4 September 2013; accepted 5 September 2013; published 23 September 2013.

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Stochastic nucleation processes and substrate abundance explain time-dependent freezing in supercooled droplets

Daniel A. Knopf, Peter A. Alpert, Assaf Zipori, Naama Reicher & Yinon Rudich

Atmos. Chem. Phys., 11, 8767–8775, 2011
www.atmos-chem-phys.net/11/8767/2011/
doi:10.5194/acp-11-8767-2011
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Atmospheric Chemistry and Physics

Heterogeneous ice nucleation: exploring the transition from stochastic to singular freezing behavior

D. Niedermeier[1], R. A. Shaw[2], S. Hartmann[1], H. Wex[1], T. Clauss[1], J. Voigtländer[1], and F. Stratmann[1]

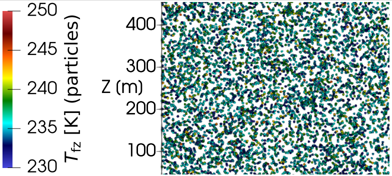
[1]Leibniz Institute for Tropospheric Research, 04318 Leipzig, Germany
[2]Dept. of Physics, Michigan Technological University, Houghton, Michigan 49931, USA

Received: 12 November 2010 – Published in Atmos. Chem. Phys. Discuss.: 28 January 2011
Revised: 24 June 2011 – Accepted: 19 August 2011 – Published: 30 August 2011

Particle-based freezing: singular (Shima et al.)

singular: INAS T_{fz} **as attribute**; initialisation by random sampling from $P(A, T_{\text{fz}})$ with lognormal A
freezing if $T_{\text{ambient}}(t) < T_{\text{fz}}|_{\text{sampled at } t=0}$

singular n_s-based (INAS)



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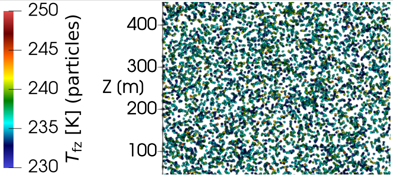

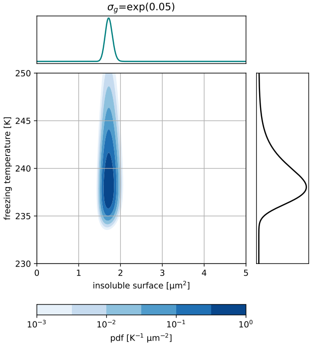

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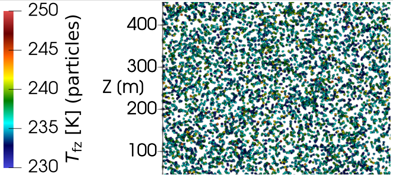

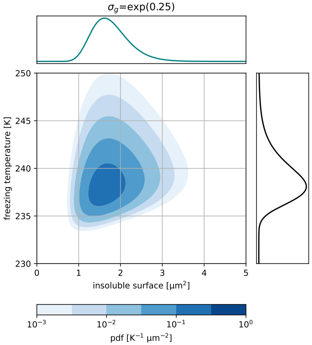

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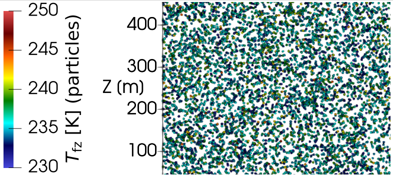

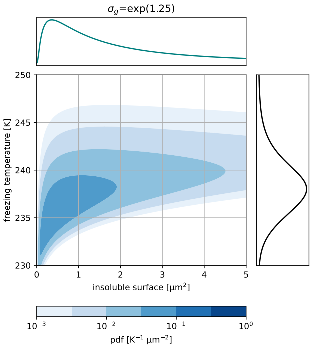

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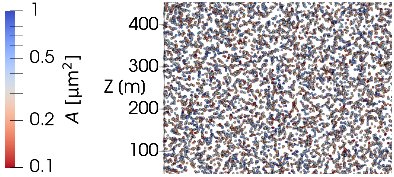
time-dependent J_{het}-based (ABIFM)



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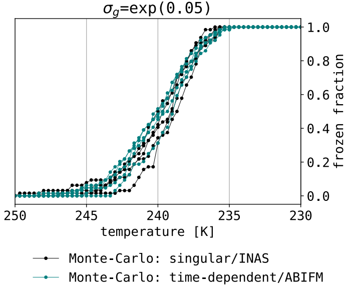

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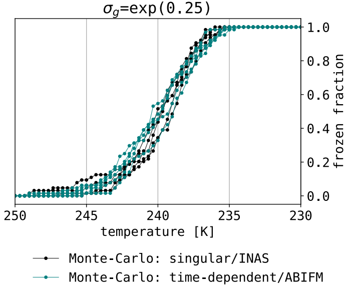

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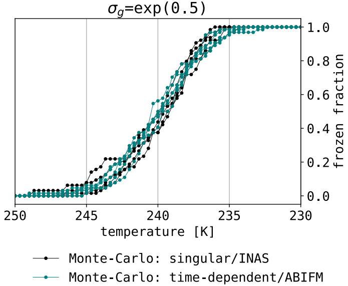

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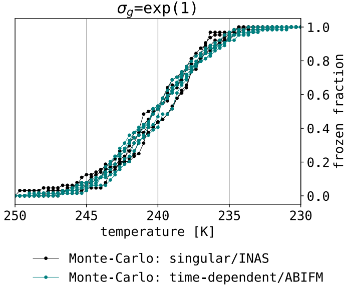

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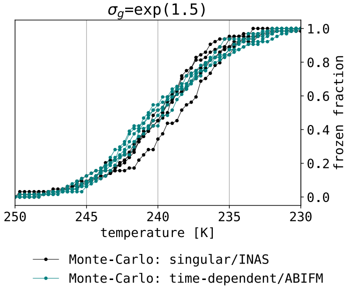

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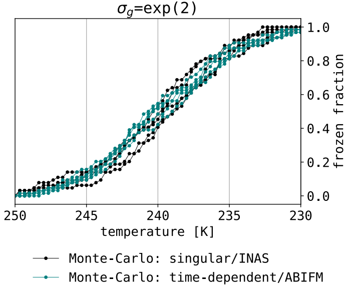

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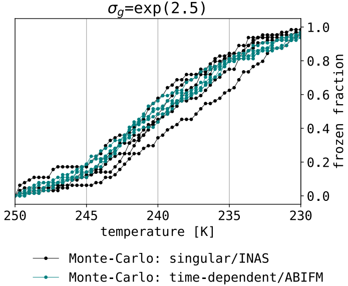

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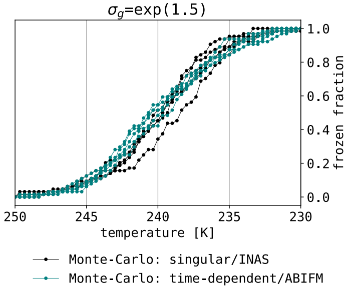

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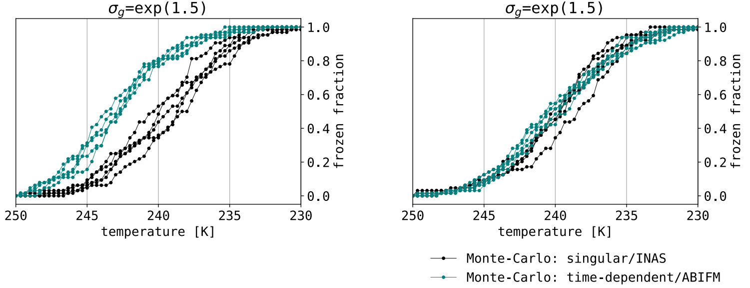

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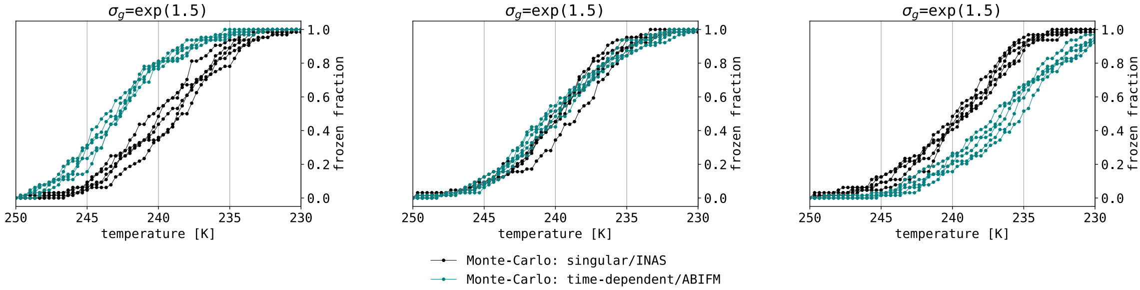

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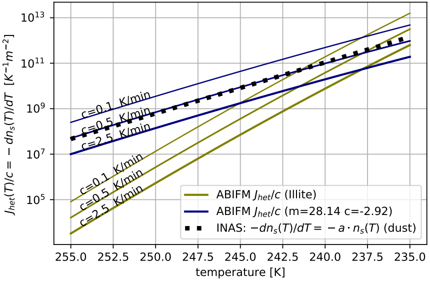


Poissonian model of freezing & Ice Nucleation Active Sites (INAS)

theory (in modern notation)
(Bigg '53, Langham & Mason '58, Carte '59, Marshall '61)

Poisson counting process with rate r:

$$P^*(\text{k events in time t}) = \frac{(rt)^k \exp(-rt)}{k!}$$

$$P(\text{one or more events in time t}) = 1 - P^*(k=0, t)$$

$$\ln(1-P) = -rt$$

introducing $J_{het}(T)$, $T(t)$ and INP surface A:

$$\ln(1-P(A,t)) = \underbrace{-A\int_0^t J_{het}(T(t'))\,dt'}_{n_s(T_{fz})}$$

INAS: $n_s(T_{fz}) = \exp(a \cdot (T_{fz} - T_{0^\circ C}) + b)$

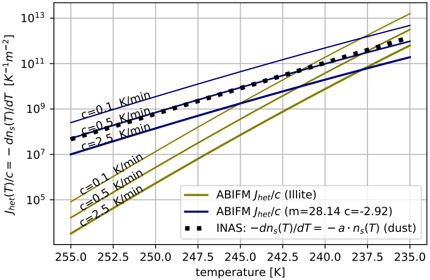
experimental $n_s(T)$ fits: e.g., Niemand et al. 2012

for a constant cooling rate $c = dT/dt$:

$$\ln(1 - P(A, t \rightsquigarrow T_{fz})) = -\frac{A}{c}\int_{T_0}^{T_0+ct} J_{het}(T')dT' = -A \cdot n_s(T_{fz})$$

$$-\frac{1}{c} J_{het}(T) = \frac{dn_s(T)}{dT} = a \cdot n_s(T)$$

experimental fits: INAS n_s (Niemand et al. '12)
ABIFM J_{het} (Knopf & Alpert '13)



cf. Vali & Stansbury '66; modified singular model (Vali '94, Murray et al. '11)
but the singular ansatz limitation of sampling T_{fz} at t=0 remains

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Is it a problem?

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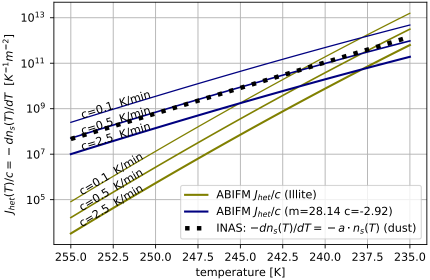

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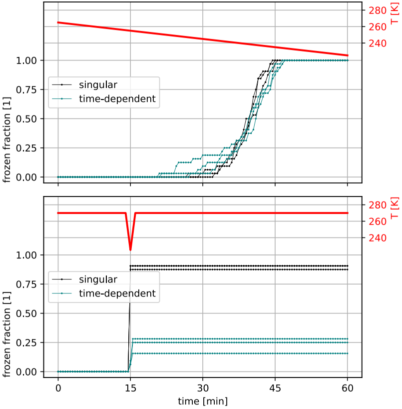
Testing different cooling-rate profiles in a box model



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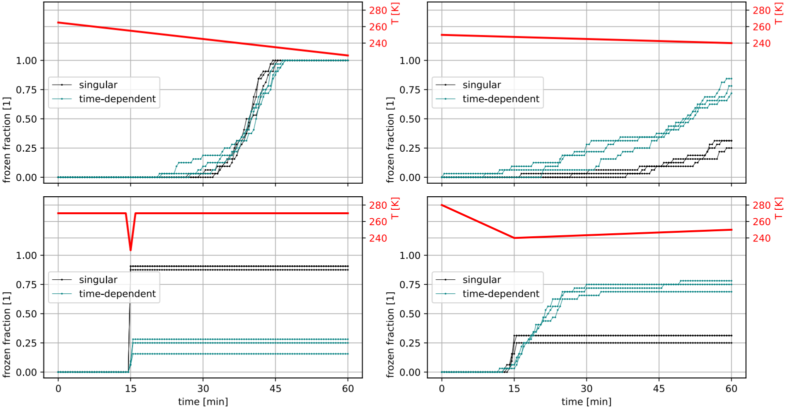

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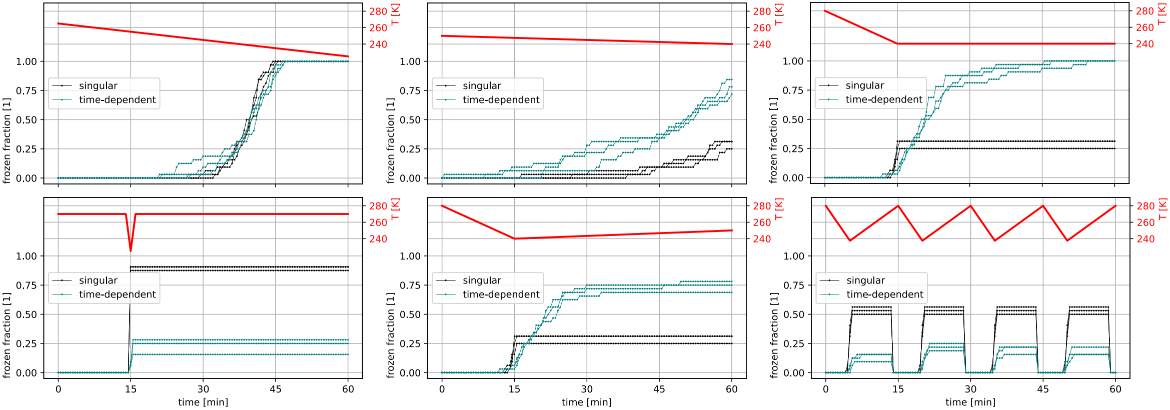
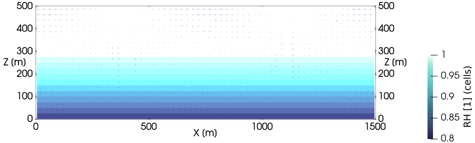


Testing three flow regimes and two immersion freezing representations

$w_{max} \approx 1/3$ m/s



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$w_{max} \approx 3$ m/s

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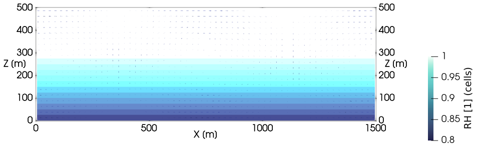

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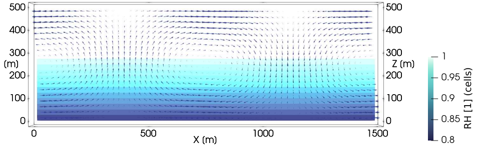

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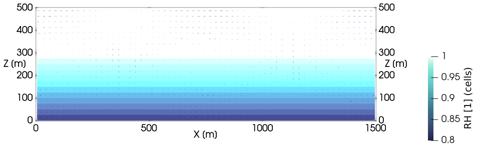

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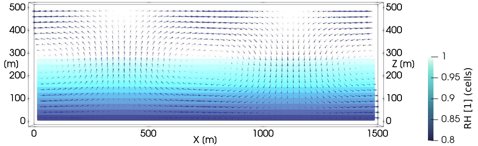

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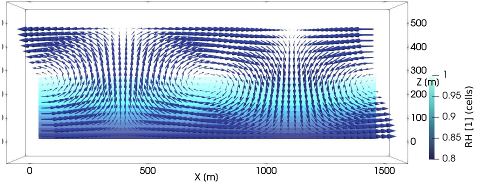

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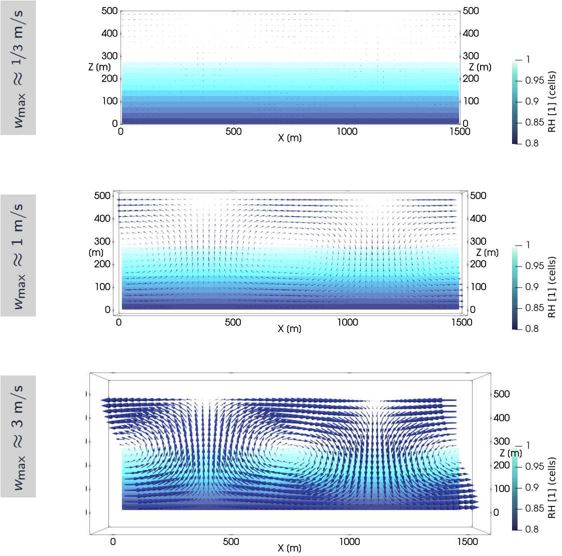
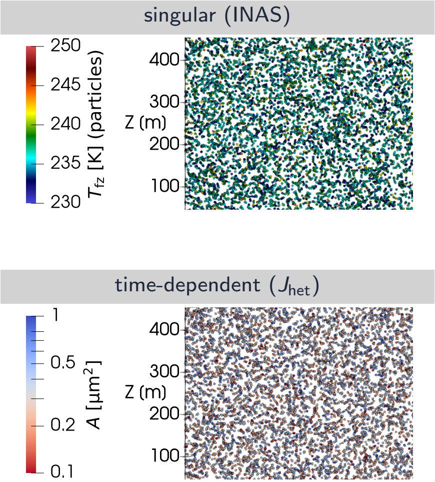
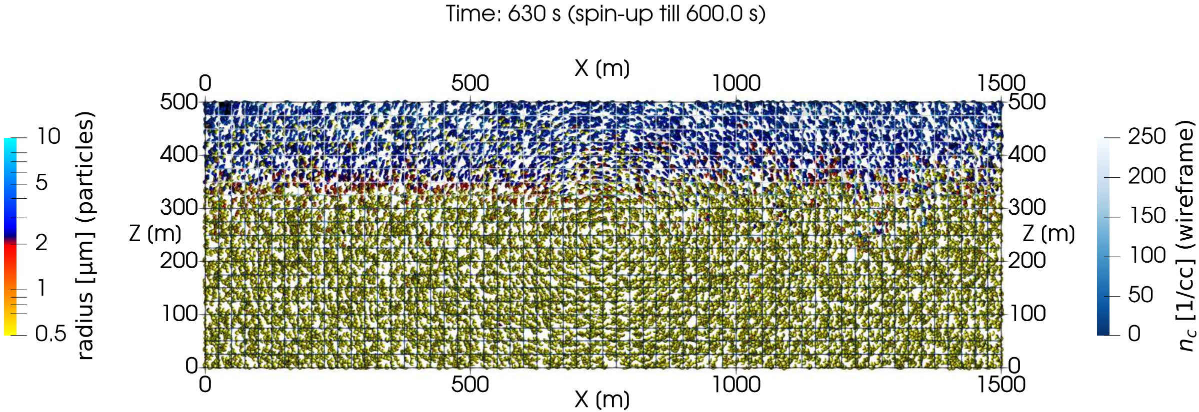




Particle-based µ-physics + prescribed-flow: glaciation



16+16 super-particles/cell for INP-rich + INP-free particles

$N_{aer} = 300/cc$ (two-mode lognormal) $N_{INP} = 150/L$ (lognormal, $D_g = 0.74$ µm, $\sigma_g = 2.55$)

spin-up = freezing off; subsequently frozen particles act as tracers

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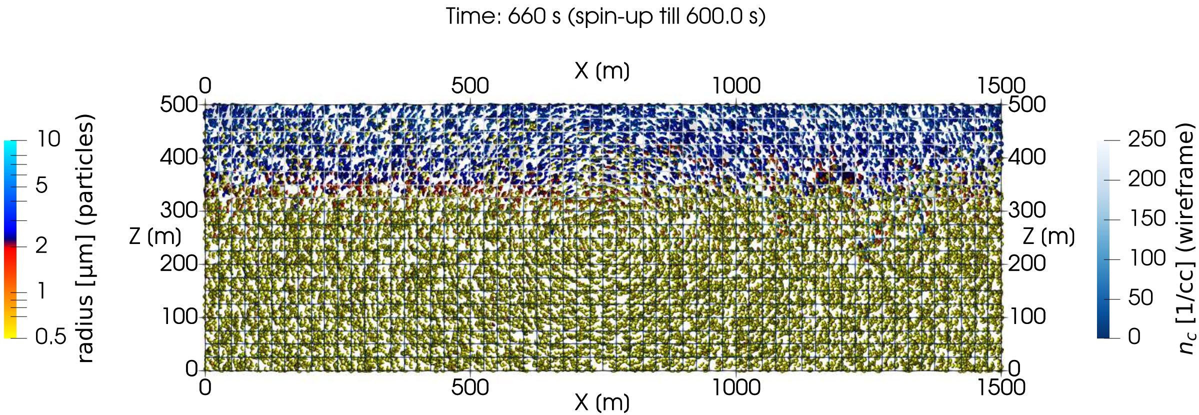

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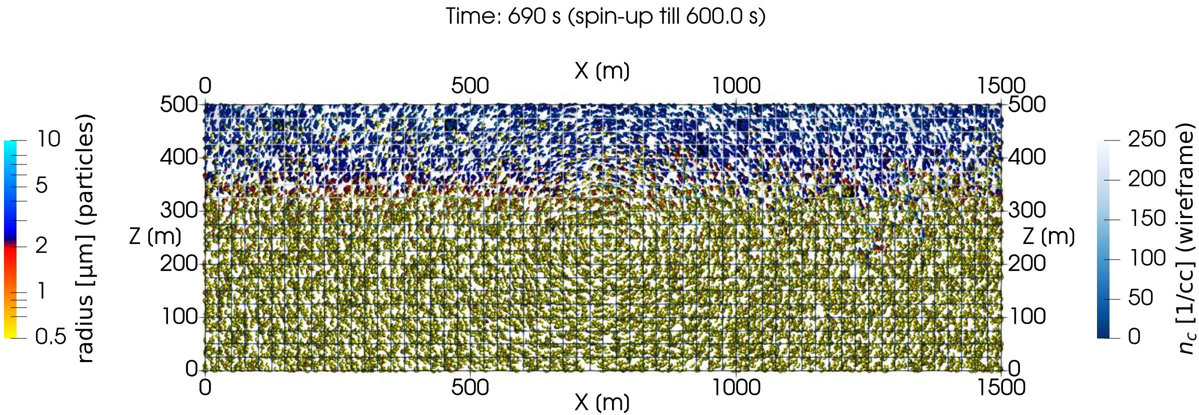

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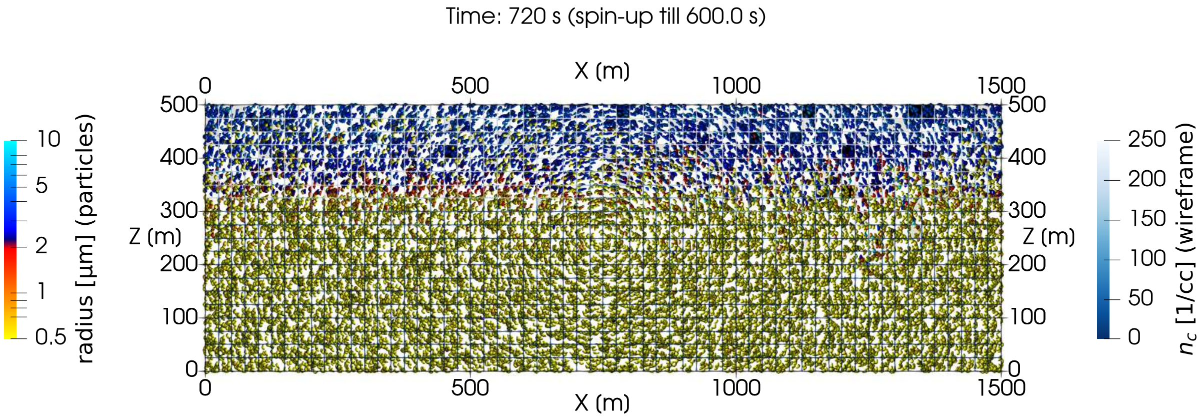

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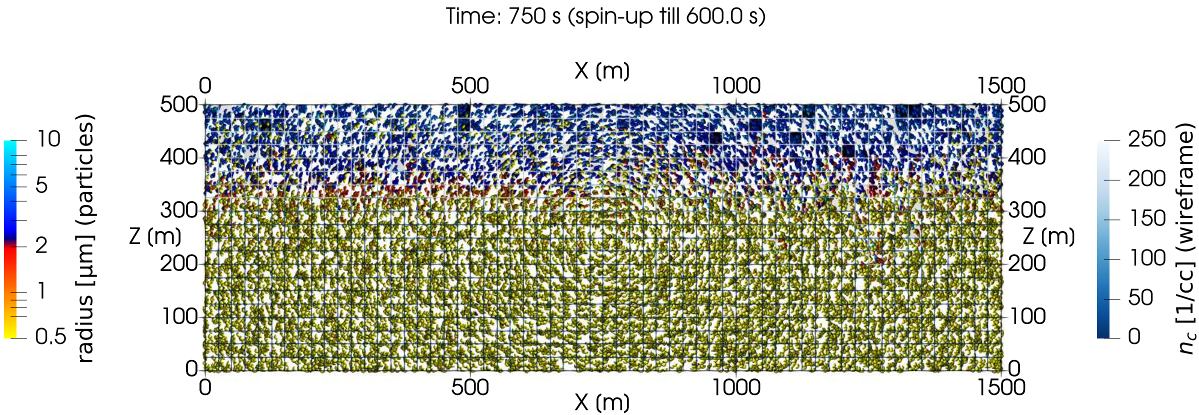

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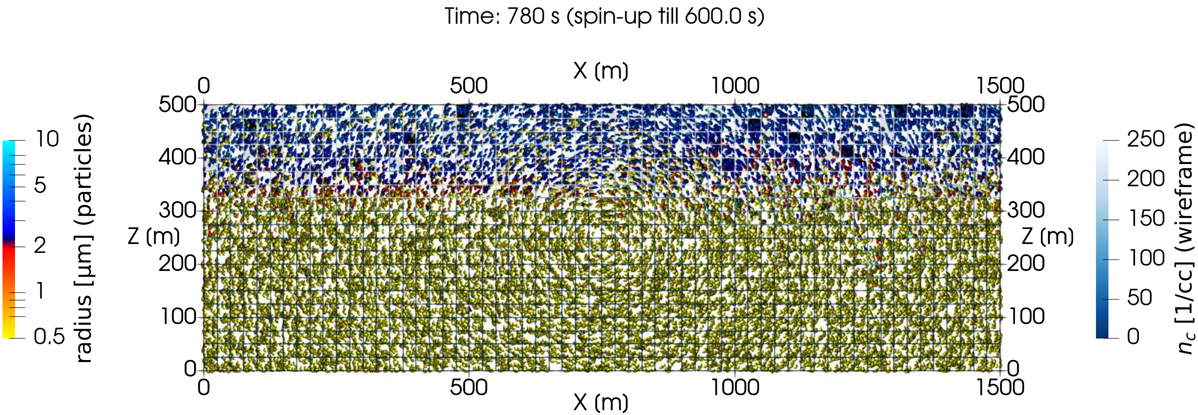

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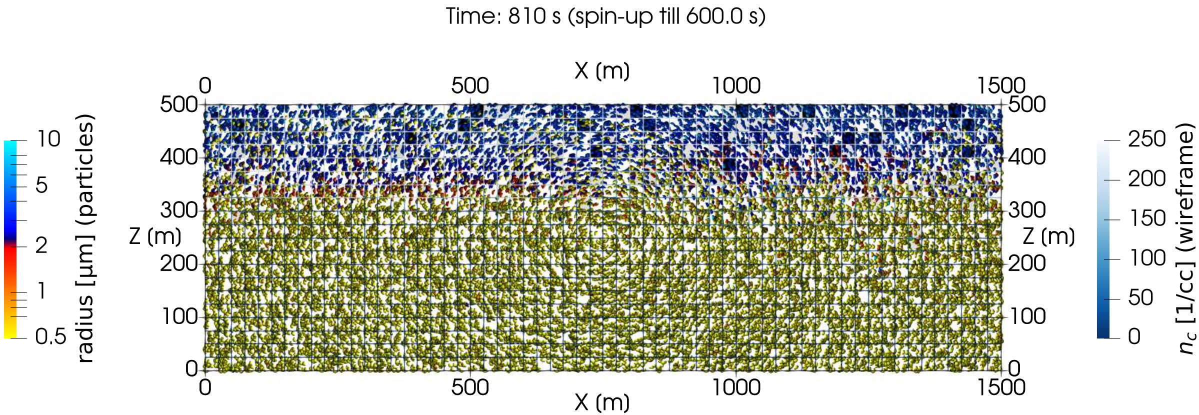

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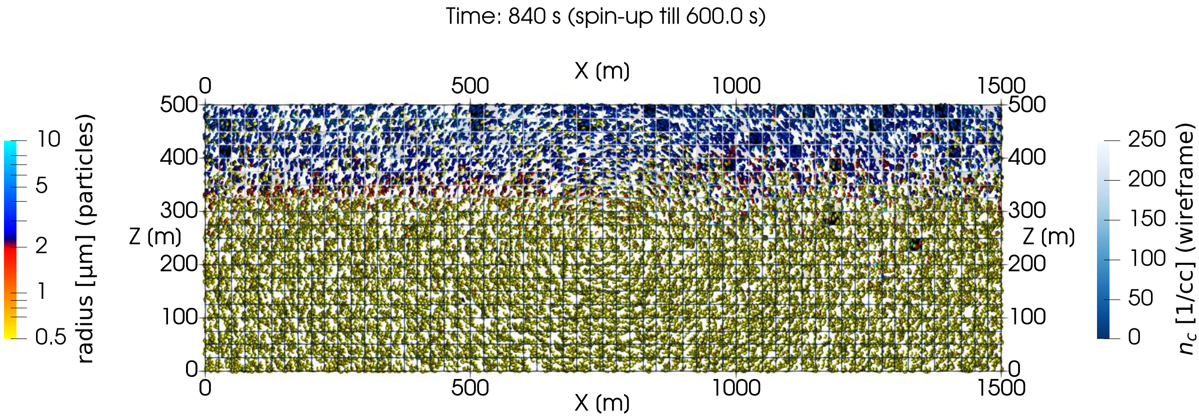

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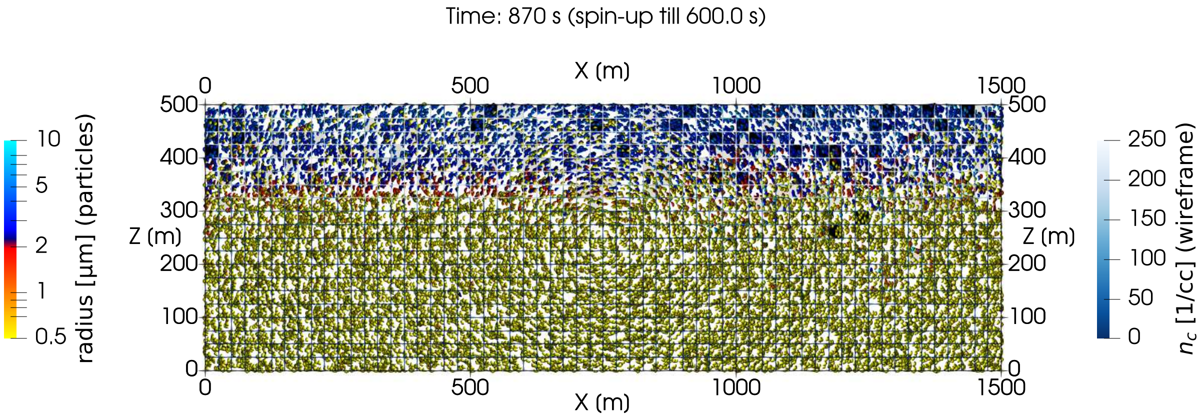

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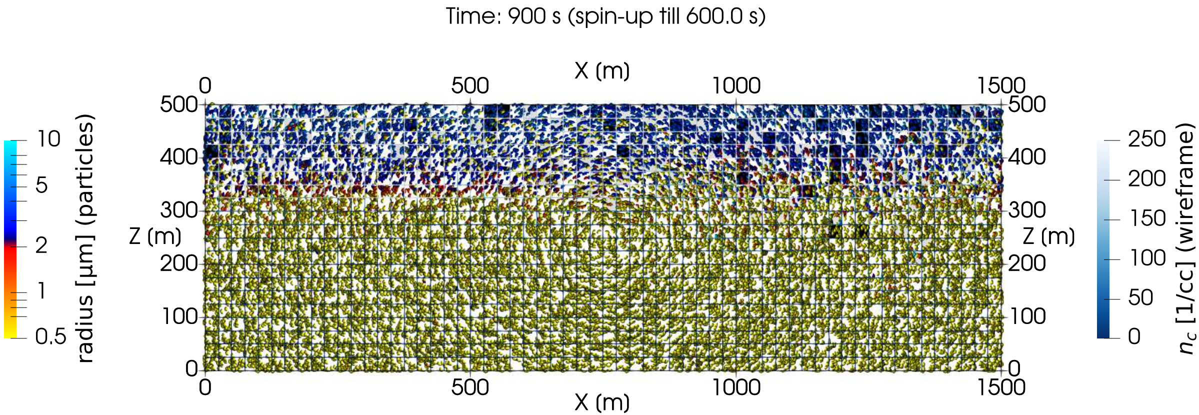

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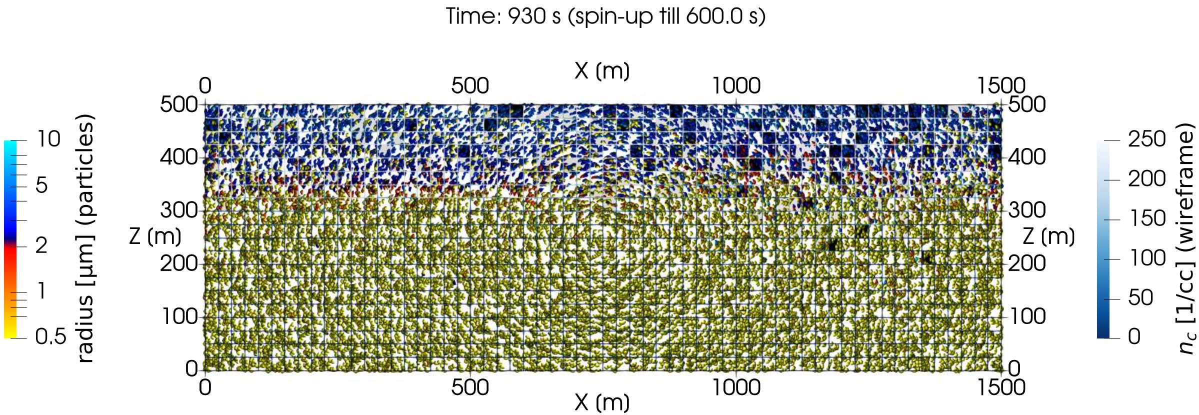

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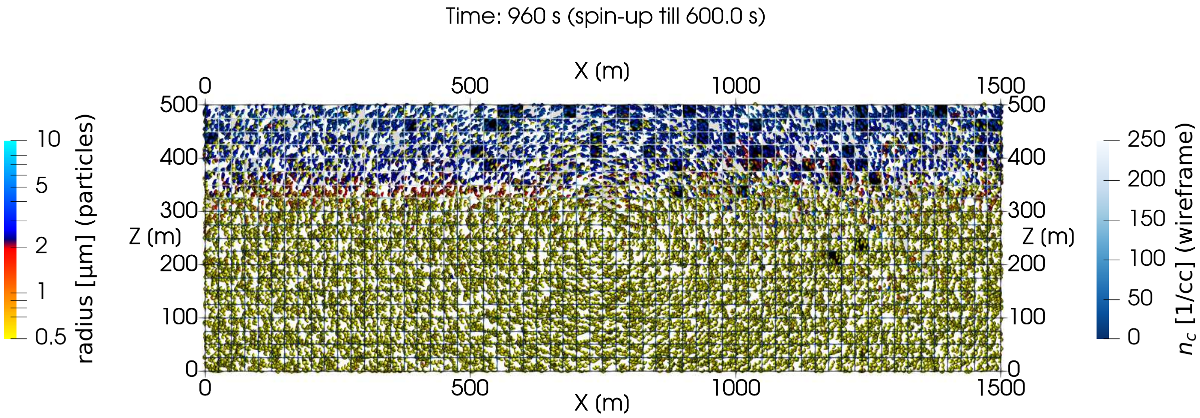

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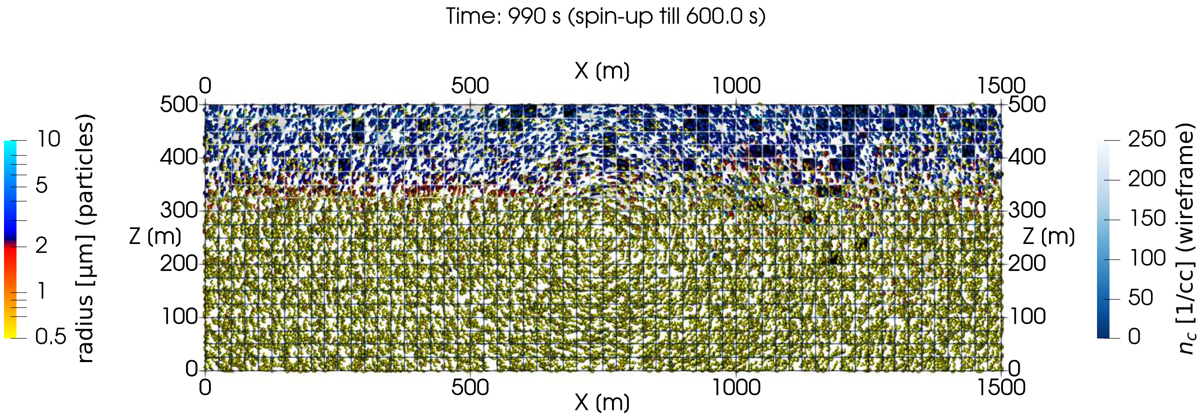

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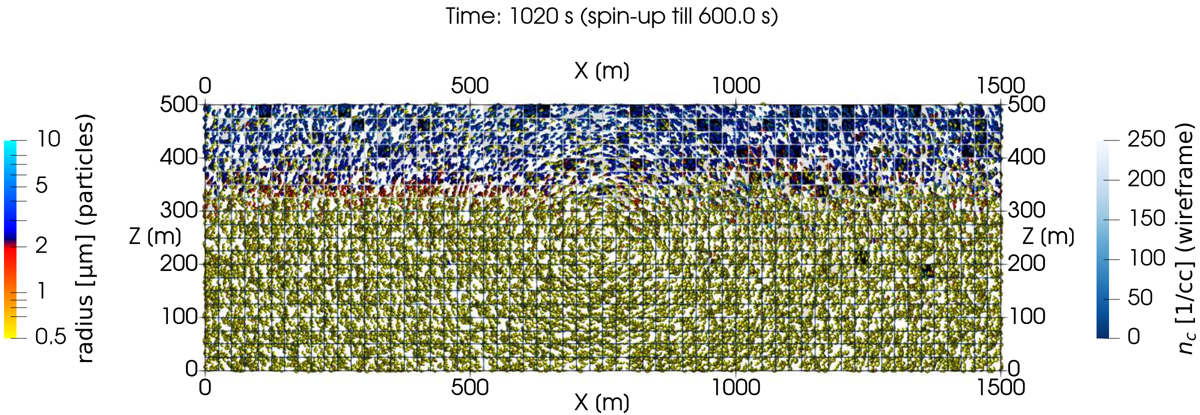

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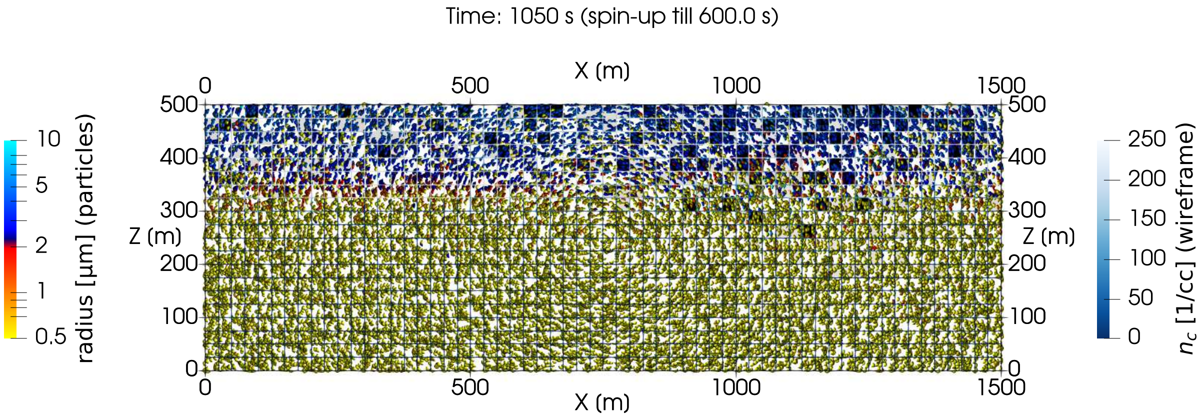

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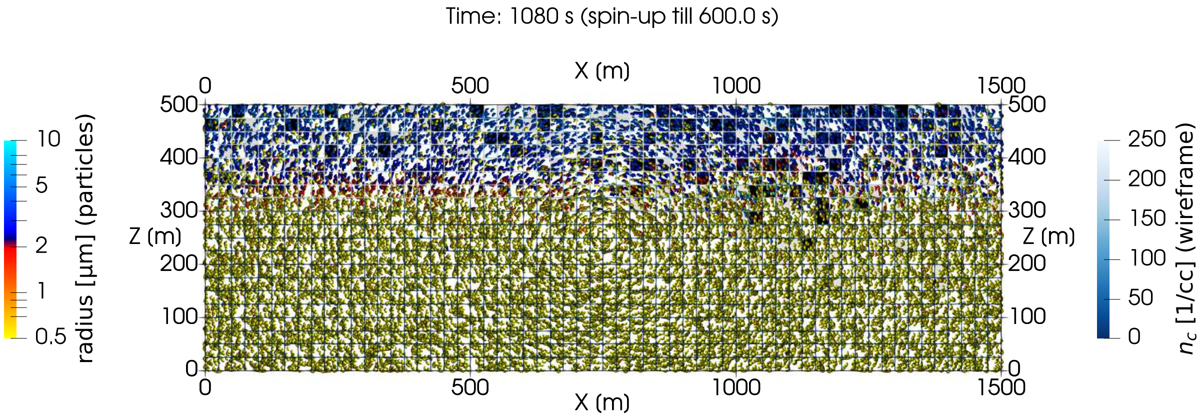

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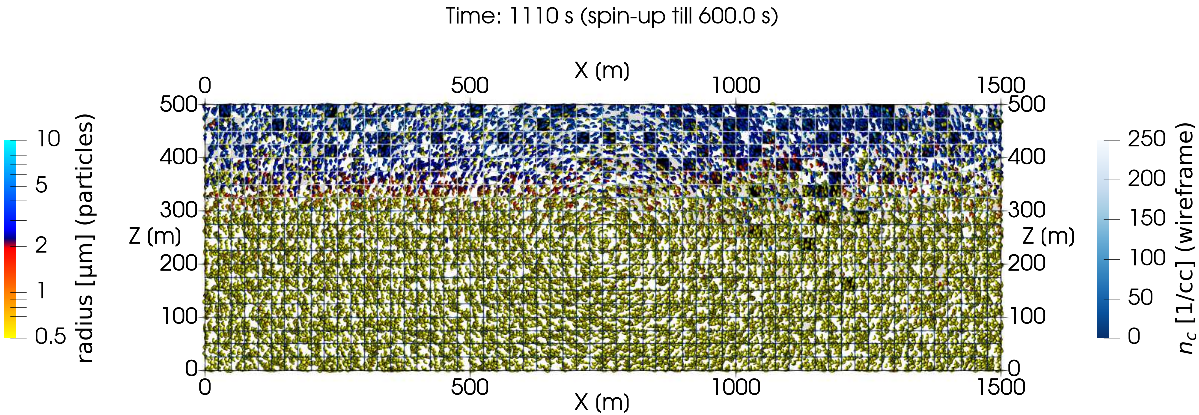

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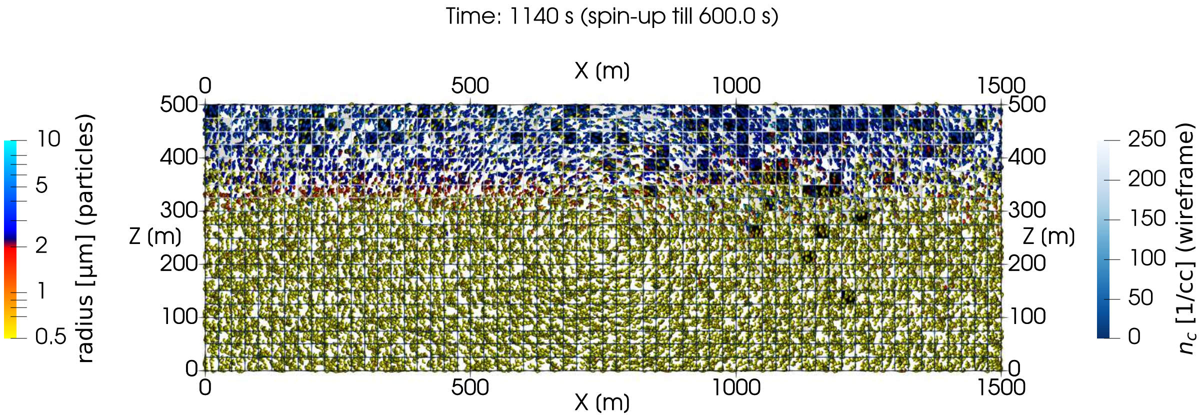

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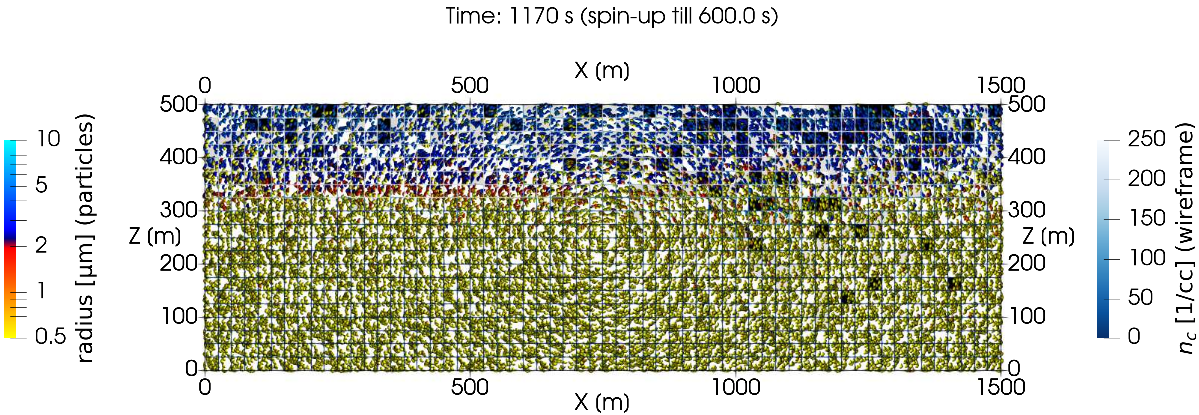

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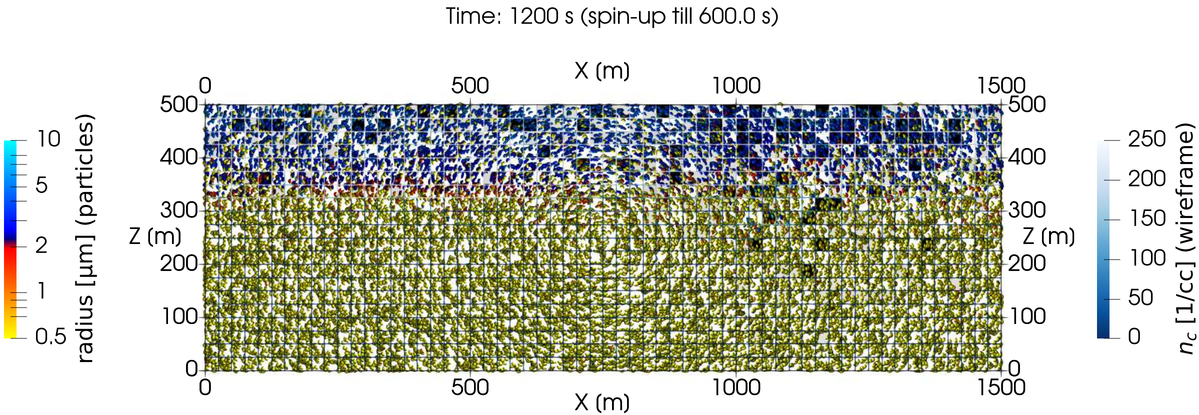

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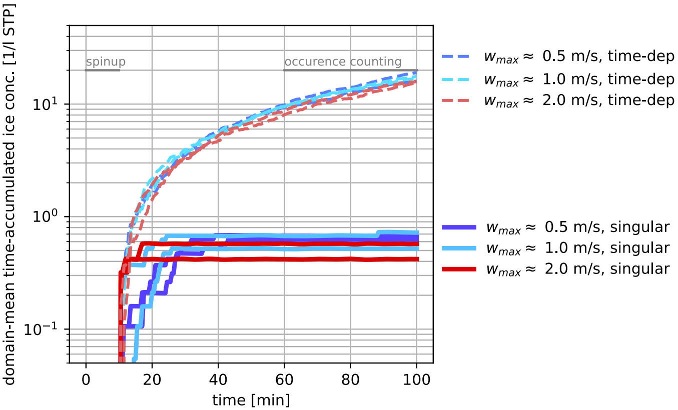

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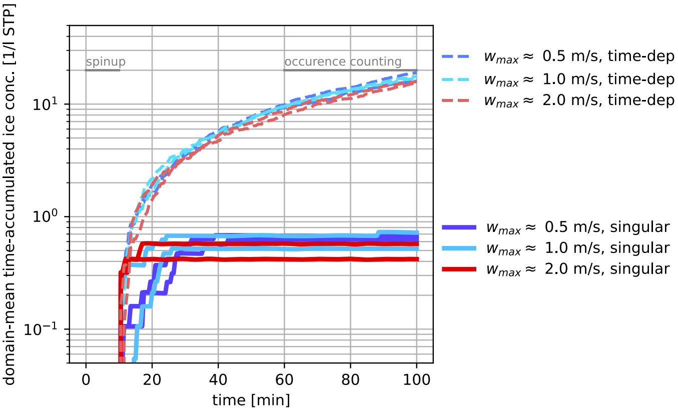

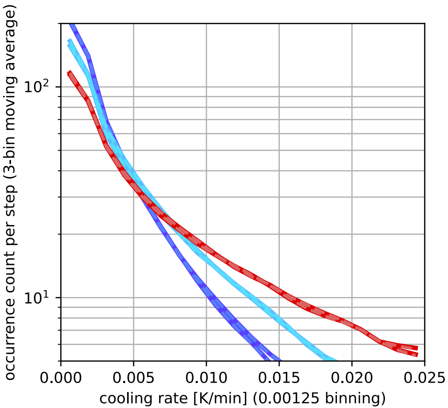

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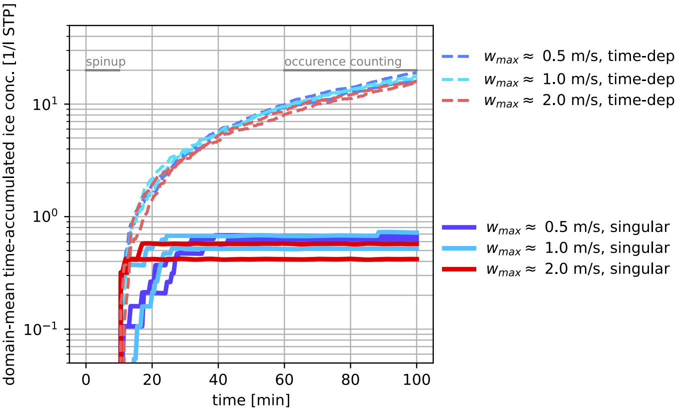

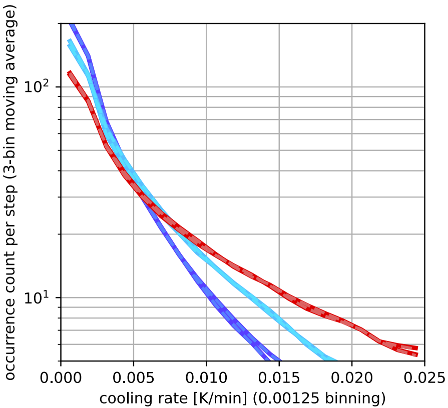

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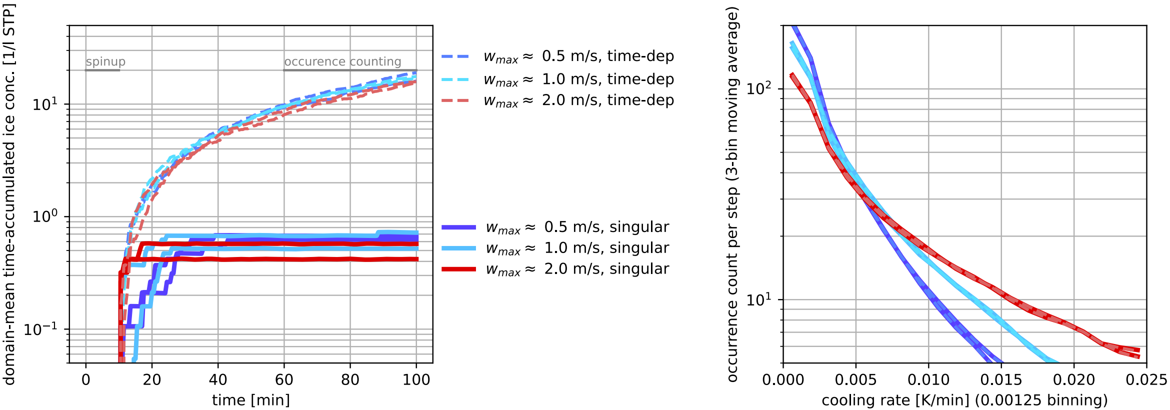

- range of cooling rates in simple flow (far from 0.5 K/min for AIDA as in Niemand et al. 2012)
- **only time-dependent scheme robust across flow regimes** (consistent with box model & theory)

100% python™ open-source code:

GitHub / OPENATMOS / PySDM

J_{het} or n_s ?

Vali 2014 (ACP)

"Interpretations of the experimental results face considerable difficulties ... two separate ways of interpreting the same observations; one assigned primacy to time the other emphasized the temperature-dependent impacts of the impurities ... dichotomy – the stochastic and singular models"

stochastic or deterministic?

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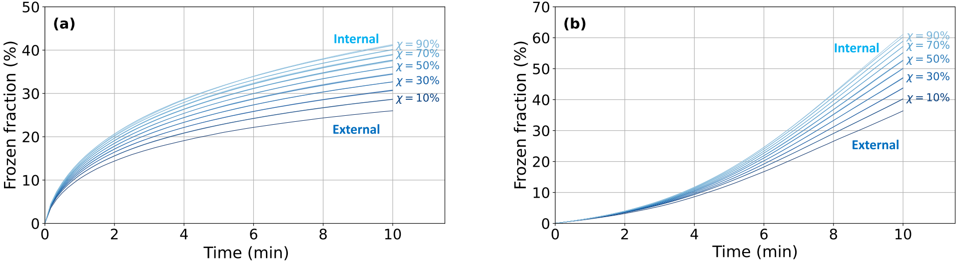
common underlying Poissonian model

The impact of aerosol mixing state on immersion freezing: Insights from classical nucleation theory and particle-resolved simulations

Wenhan Tang , Sylwester Arabas , Jeffrey H. Curtis , Daniel A. Knopf , Matthew West , and Nicole Riemer

Abstract. Immersion freezing, initiated by ice-nucleating particles (INPs) in supercooled aqueous droplets, plays an important role in the formation of ice crystals within clouds. The efficiency of immersion freezing depends strongly on INP composition and, crucially, on the mixing state—how chemical species are distributed across the particle population. Here, we quantify the impact of aerosol mixing state on immersion freezing using a combined theoretical and particle-resolved modeling approach.



(a): isothermal freezing conditions with −20 °C
(b): constant cooling rate from −10 °C to −30 °C within 10 minutes

Thank you for your attention!

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Immersion Freezing in Particle-Based Aerosol-Cloud Microphysics: A Probabilistic Perspective on Singular and Time-Dependent Models

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