

Immersion freezing in particle-based cloud μ -physics models

S. Arabas¹, J.H. Curtis², I. Silber³, A. Fridlind⁴, D.A. Knopf⁵, M. West² & N. Riemer²



funding:



Mini-workshop on particle-based cloud modeling 2025, Kobe, Sep 26th 2025

Immersion freezing: bacteria and the Olympics



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

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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

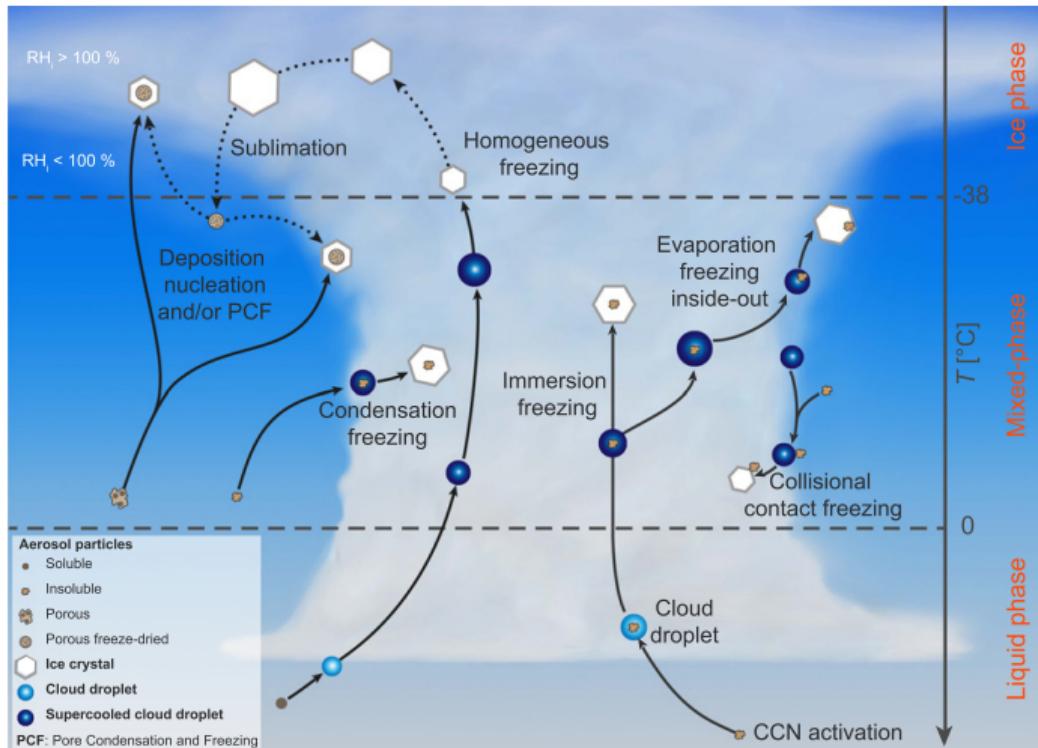
The unstable ice nucleation properties of Snomax® bacterial particles

Michael Polen¹, Emily Lawlis¹, and Ryan C. Sullivan¹

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

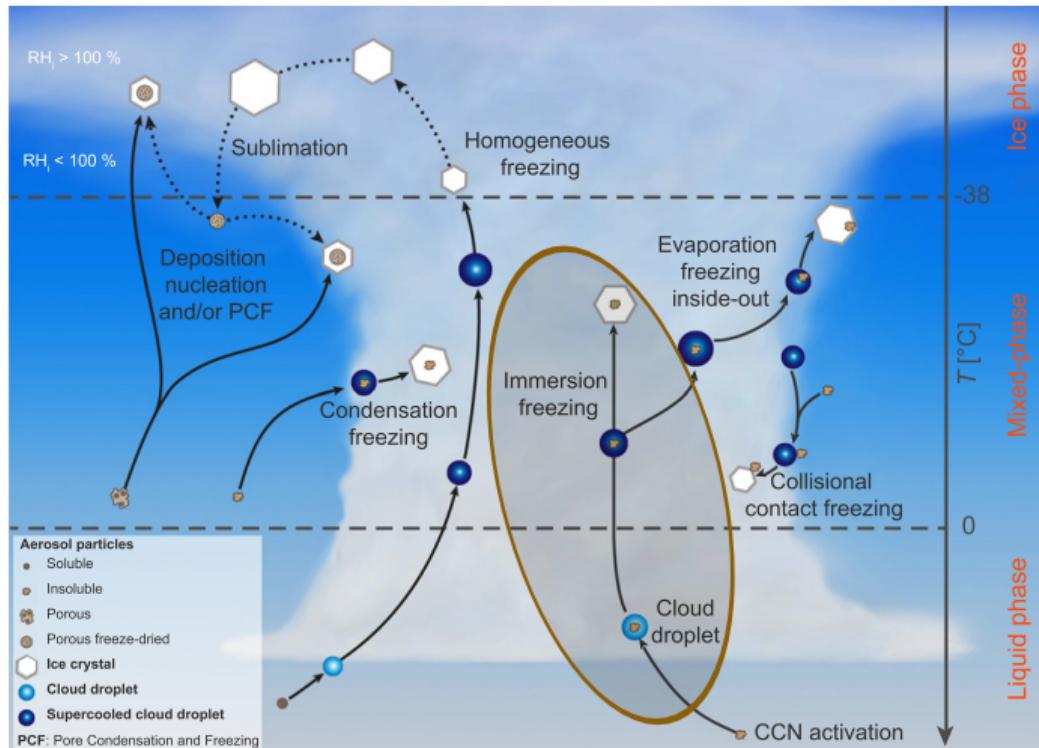
Abstract Snomax® is often used as a surrogate for biological ice nucleating particles (INPs) and has recently been proposed as an INP standard for evaluating ice nucleation methods. We have found the immersion freezing properties of Snomax particles to be substantially unstable, observing a loss of ice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

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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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 :

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

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

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

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

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

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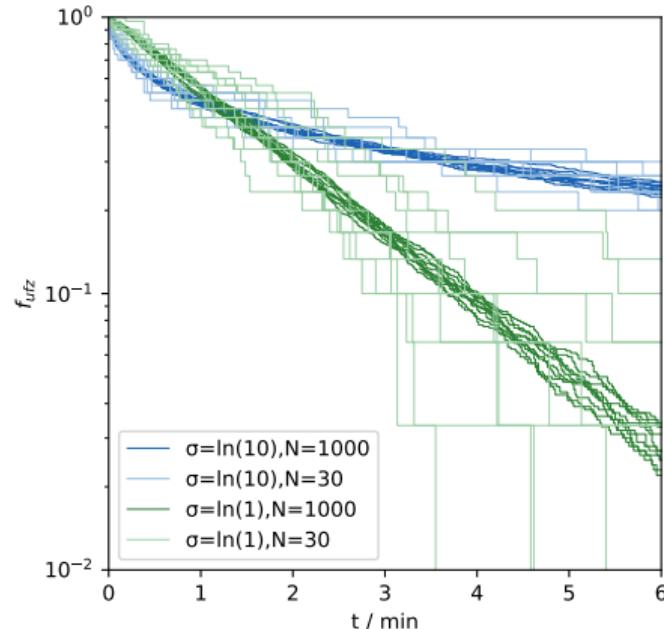
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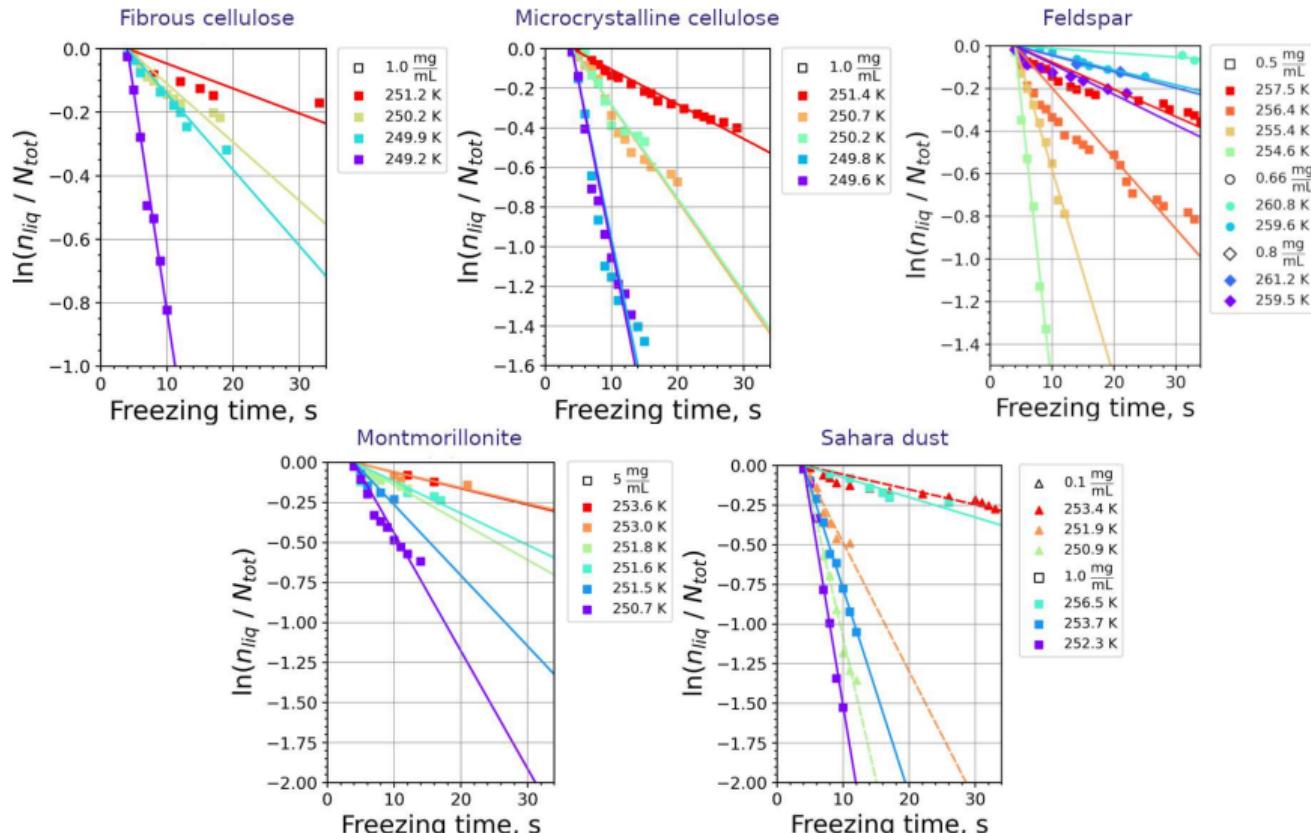
$$\ln(1 - P(A, t)) = -A \int_0^t J_{\text{het}}(T(t')) dt'$$

Monte Carlo: const J_{het} , lognormal A



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

Szakáll et al. 2021, ACP 21: isothermal experiments (IPA, Mainz)



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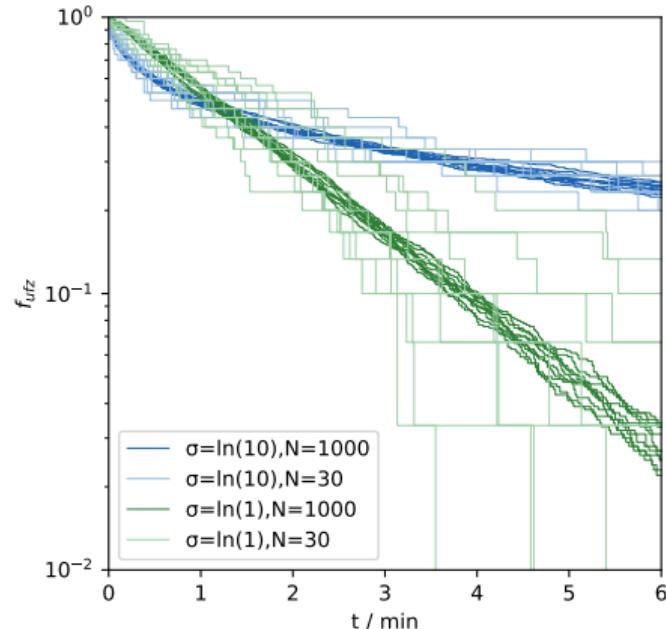
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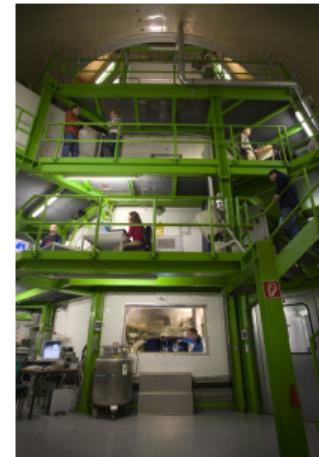
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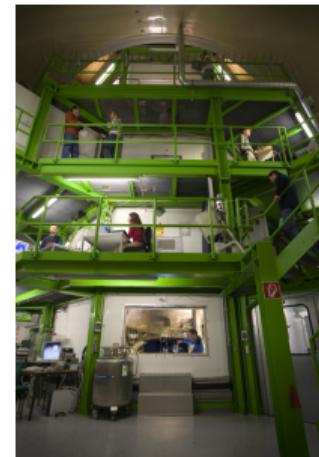
experimental $n_s(T)$ fits: e.g., Niemand et al. 2012

AIDA @ KIT



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

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AIDA cooling rate: ca. $0.5\text{ K}/\text{min}$

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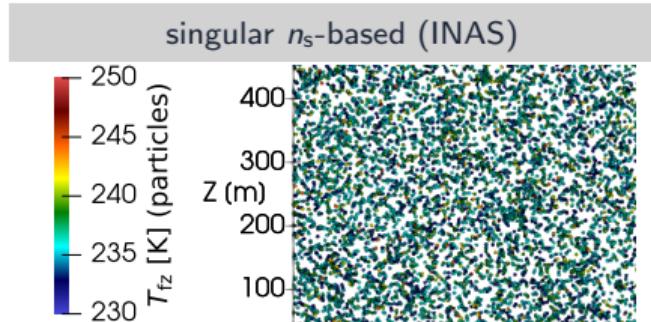
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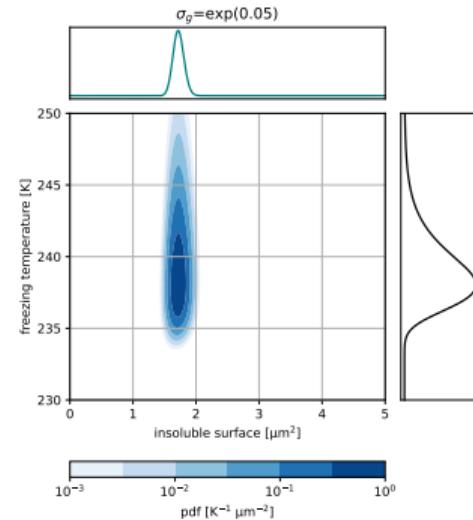
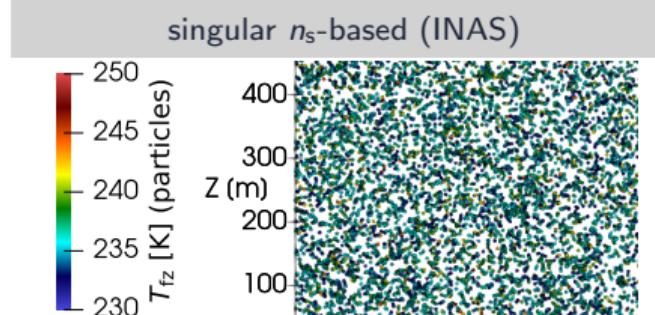
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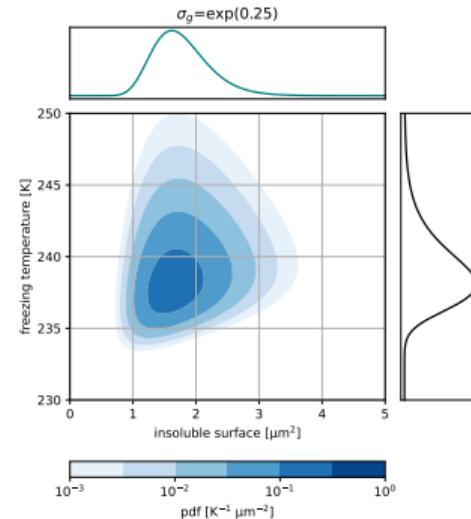
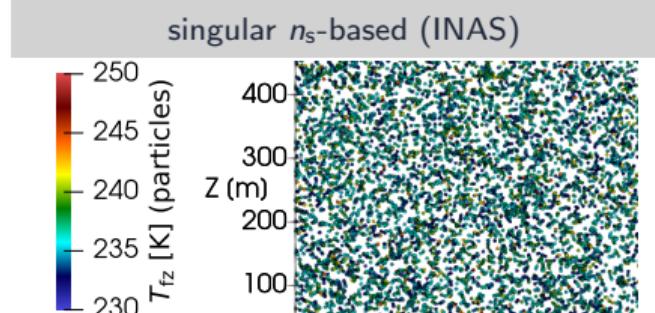
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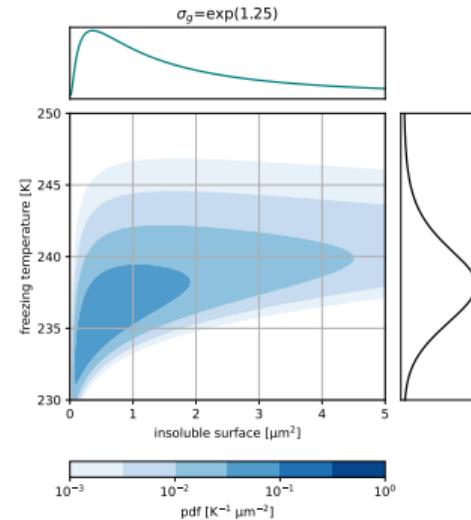
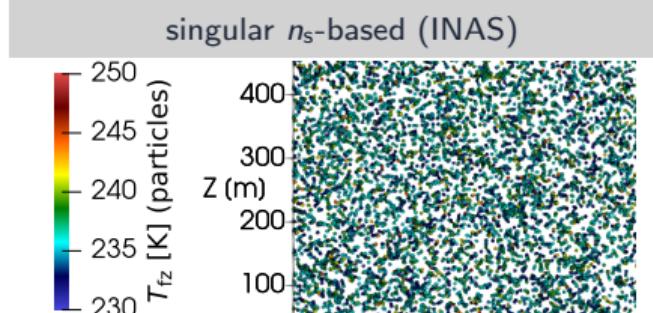
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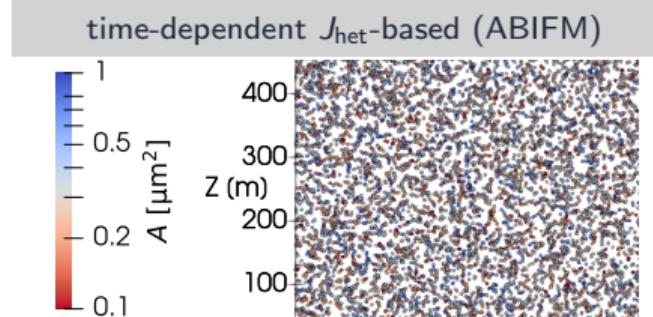
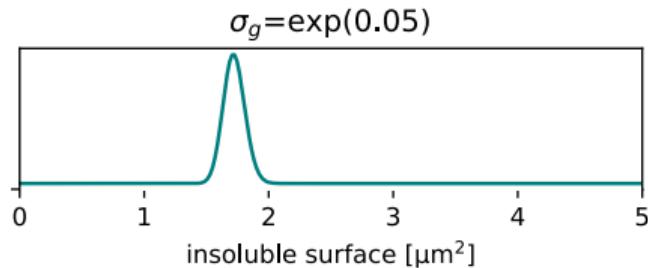
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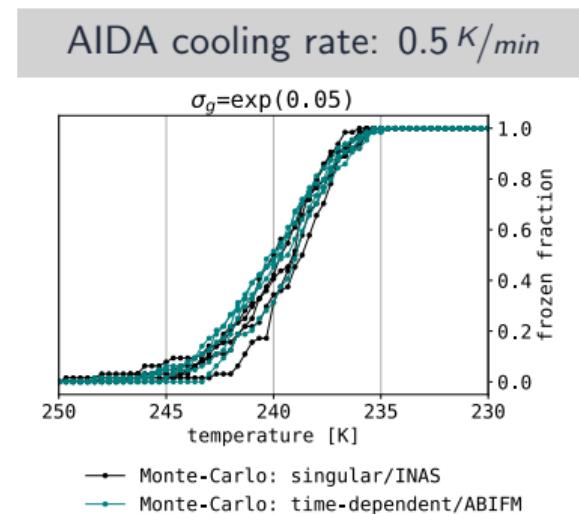
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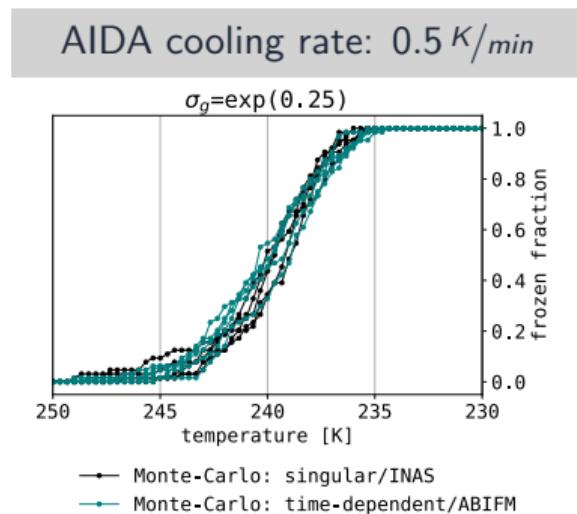
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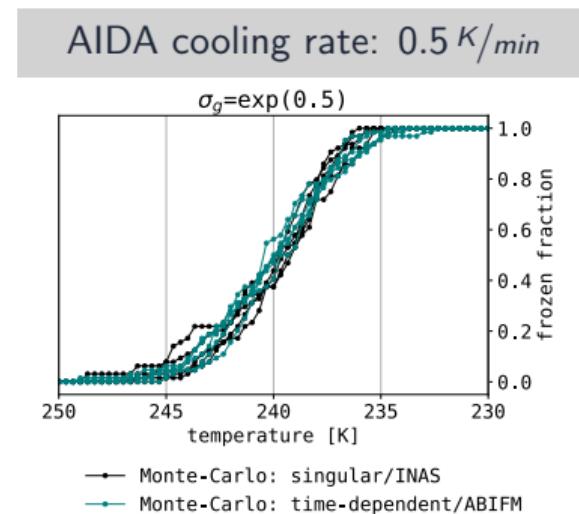
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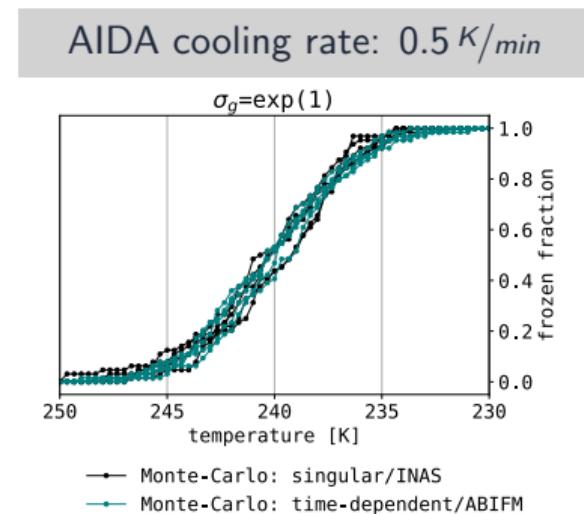
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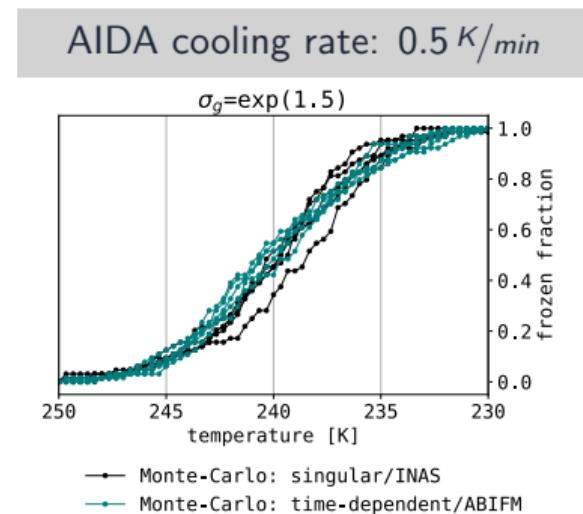
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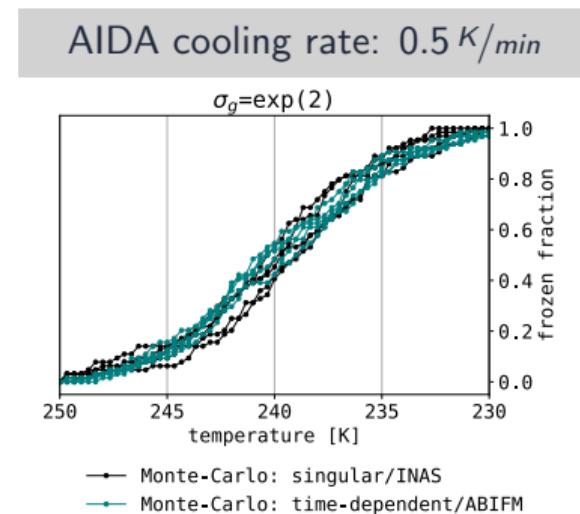
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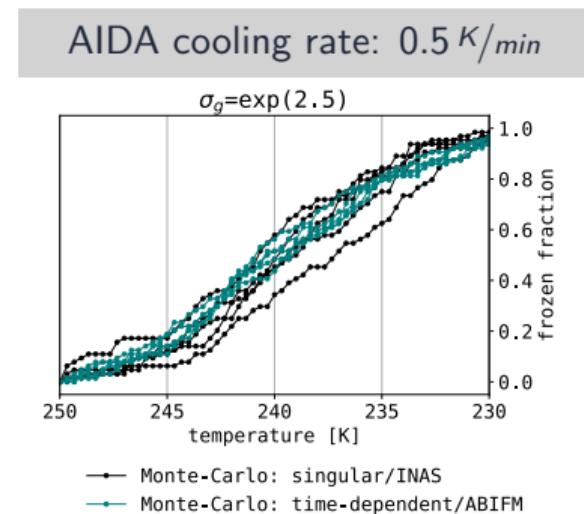
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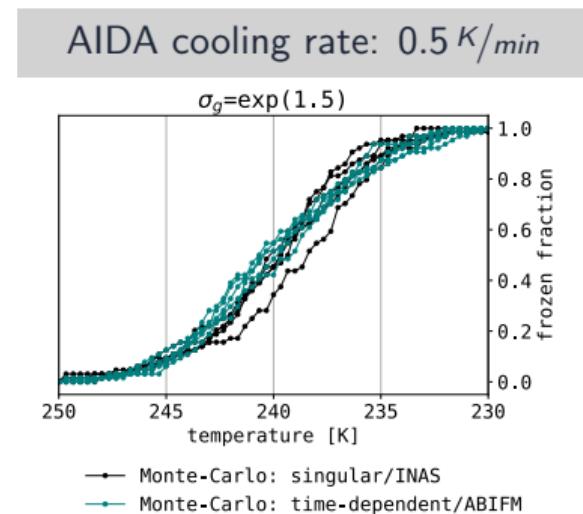
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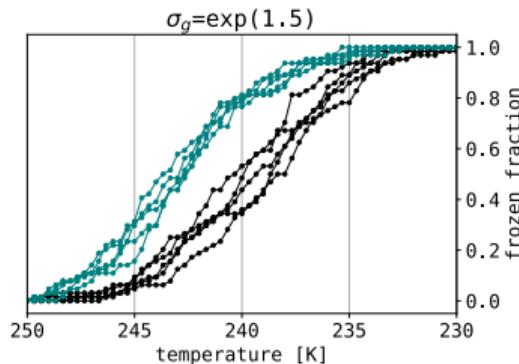


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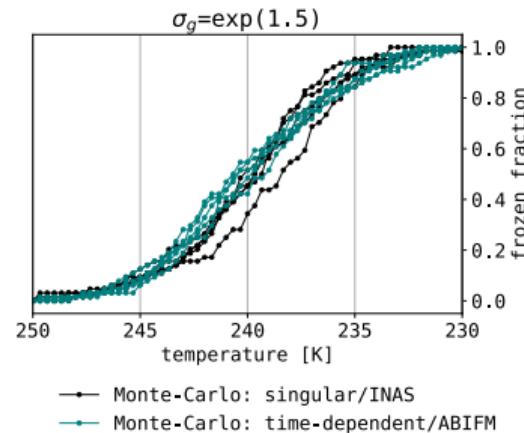
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AIDA cooling rate: 0.5 K/min

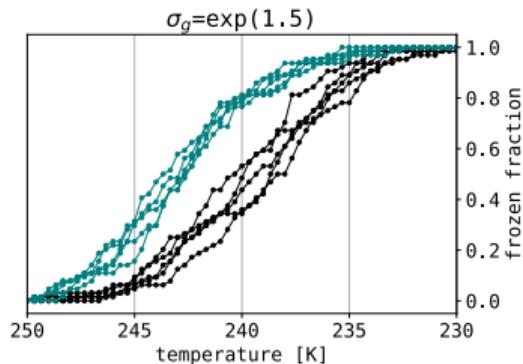


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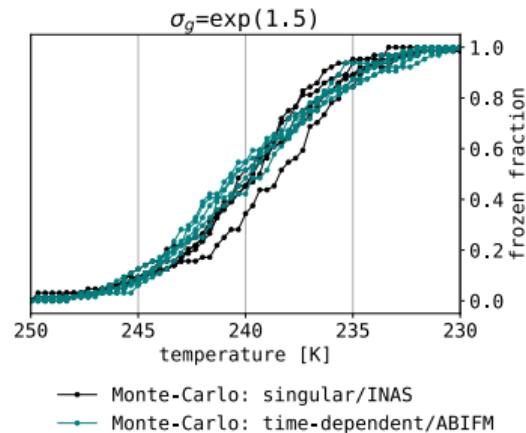
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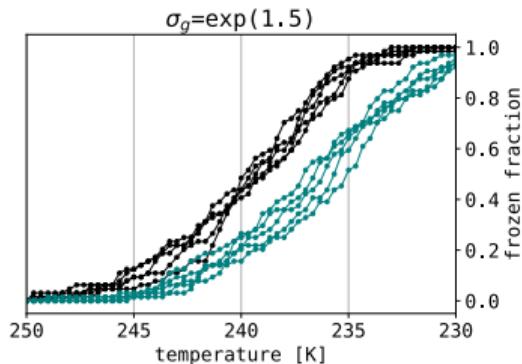
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Knopf & Alpert '13

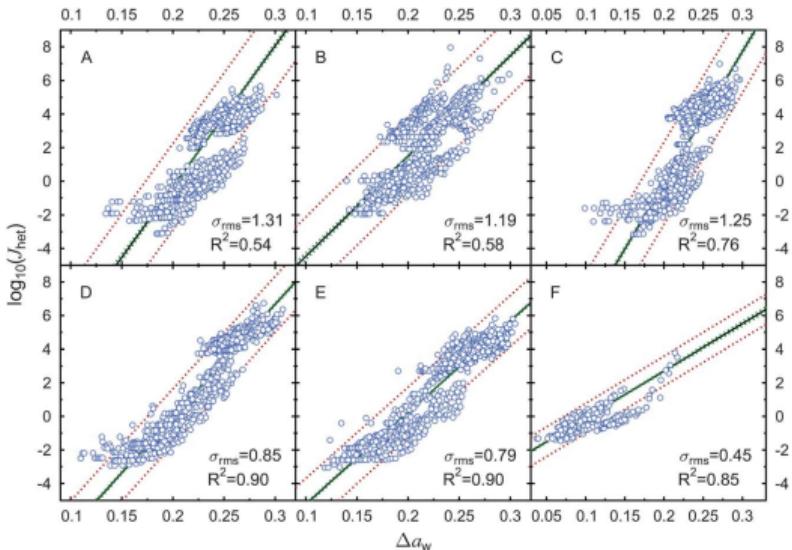


Fig. 3 The decadal log of the heterogeneous ice nucleation rate coefficients, $\log_{10}U_{\text{het}}$, are shown as a function of Δa_w for individually analysed freezing events, initiated by the different IN types investigated in this study and previous work.^{41,43,57,66,73,75,78,95} $\log_{10}U_{\text{het}}$ are shown for (A) *Nannochloris atomus*, (B) *Thalassiosira pseudonana*, (C) Pahokee Peat, (D) Leonardite, (E) Illite, and (F) 1-nonadecanol. The solid black line is a linear fit where dashed green and red lines represent confidence intervals and prediction bands at 95% level. The root mean square error, σ_{rms} , and the adjusted coefficient of determination, R^2 , are given in each panel.

Kanji et al. '17

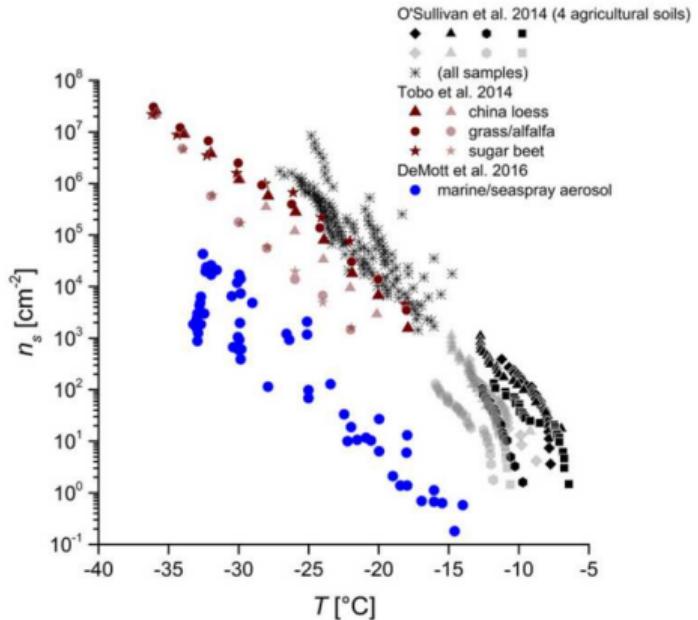


FIG. 1-6. Ice nucleation active site densities n_s as a function of temperature for H_2O_2 (hydrogen peroxide) treated (lighter-shaded symbols) and untreated (dark symbols) agricultural soil dusts in comparison to the n_s of marine aerosol. Differences between various black symbols are for organic content (OC). High OC (12.7 wt%)

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

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

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 :

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

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

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

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

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

$\overbrace{\hspace{10em}}$
 $n_s(T_{\text{fz}})$

$$\text{INAS: } n_s(T_{\text{fz}}) = \exp(a \cdot (T_{\text{fz}} - T_0 \cdot c) + b)$$

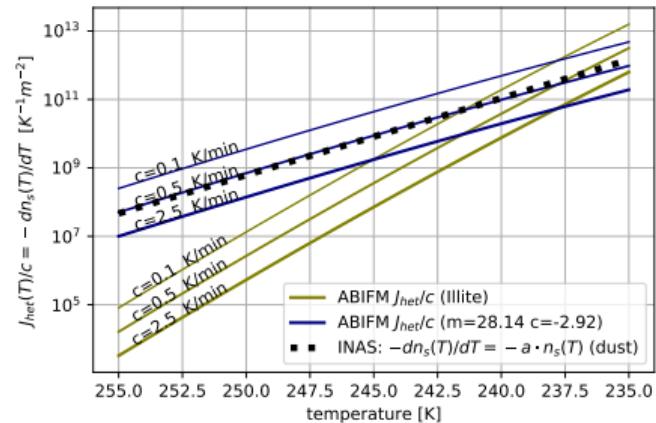
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experimental fits: INAS n_s (Niemand et al. '12)
ABIFM J_{het} (Knopf & Alpert '13)



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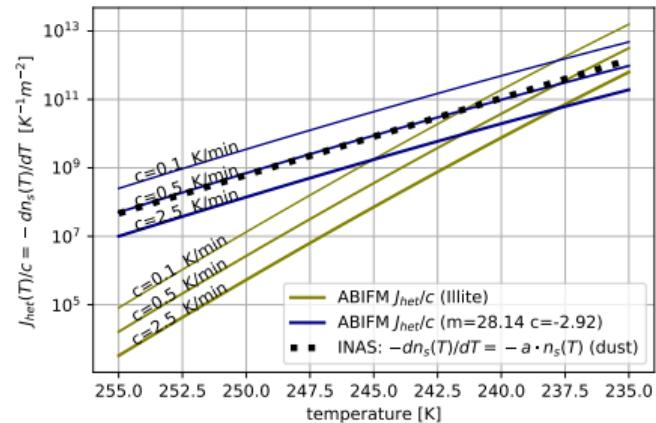
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experimental fits: INAS n_s (Niemand et al. '12)
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Poissonian model of freezing & Ice Nucleation Active Sites (INAS)

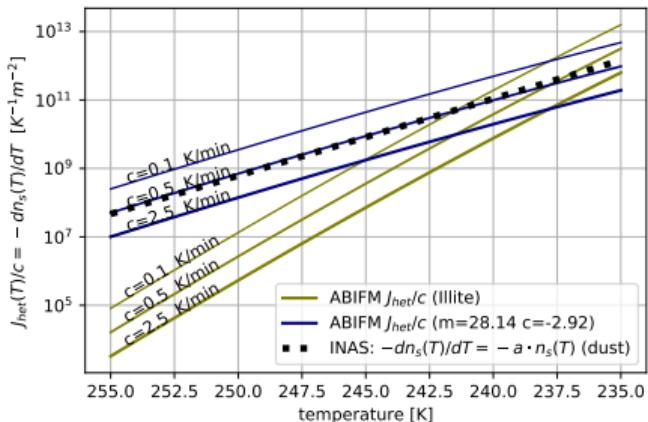
for a constant cooling rate $c = dT/dt$:

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

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experimental fits: INAS n_s (Niemand et al. '12)
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Is it a problem?



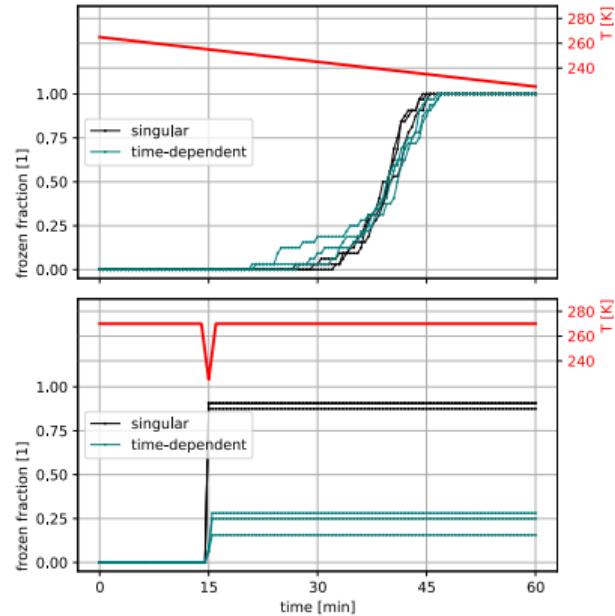


Immersion Freezing in Particle-Based Aerosol-Cloud Microphysics: A Probabilistic Perspective on Singular and Time-Dependent Models

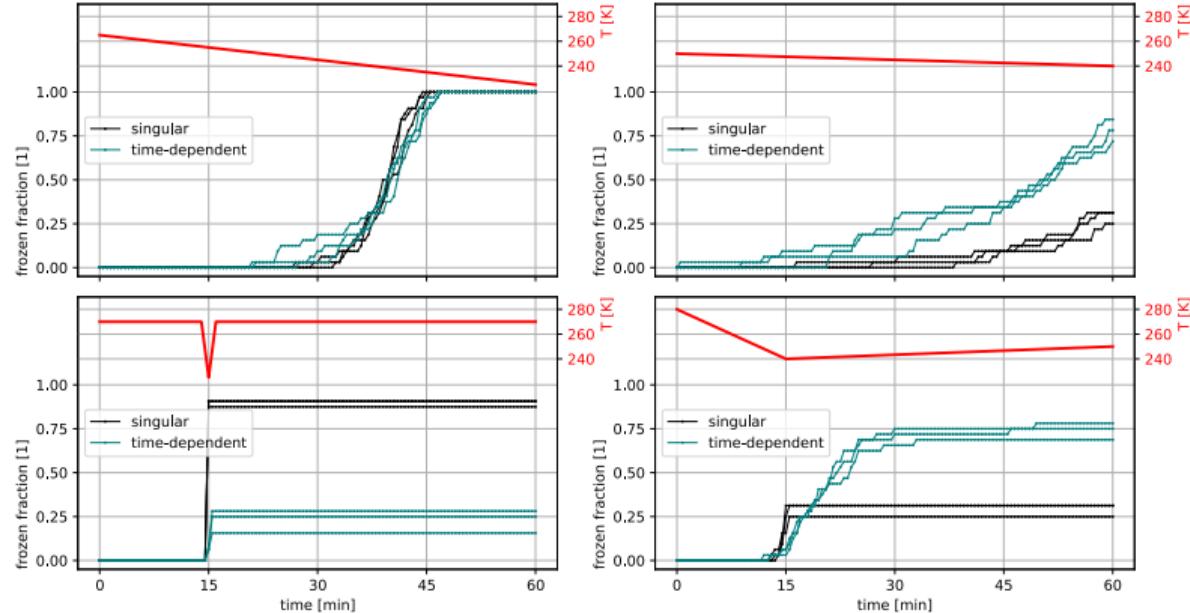
Sylwester Arabas¹ , Jeffrey H. Curtis² , Israel Silber^{3,4} , Ann M. Fridlind⁵ ,
Daniel A. Knopf⁶ , Matthew West⁷ , and Nicole Riemer²

10.1029/2024MS004770

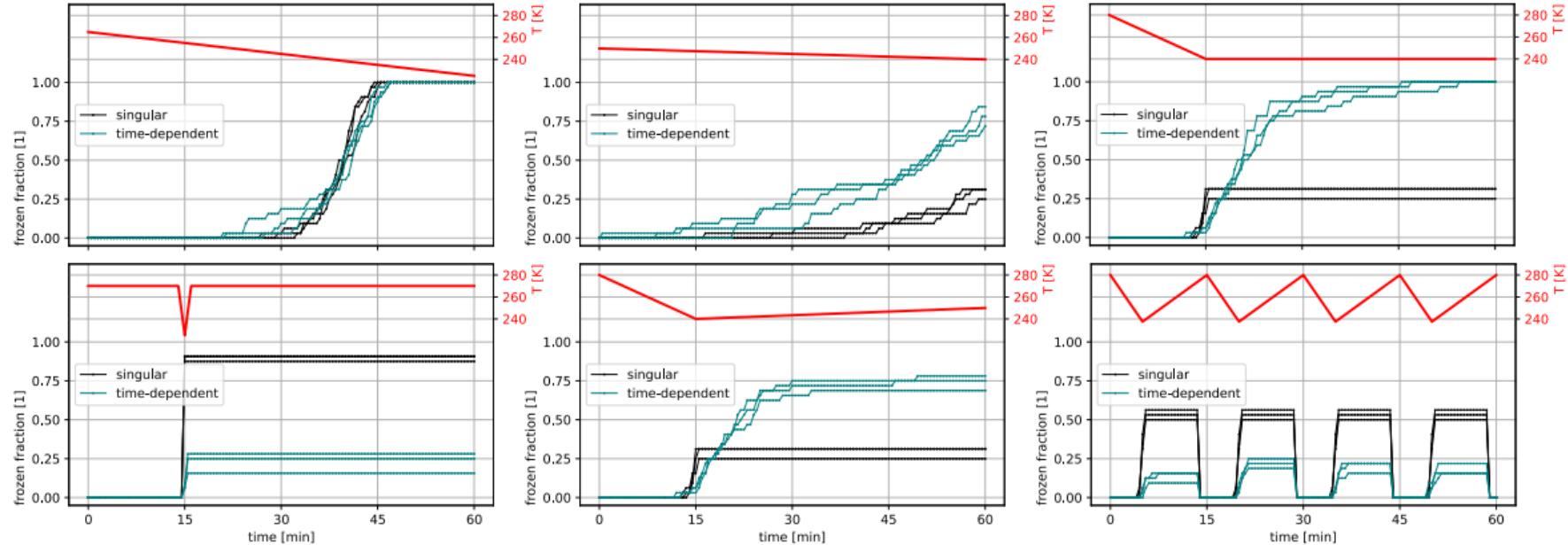
Testing different cooling-rate profiles in a box model



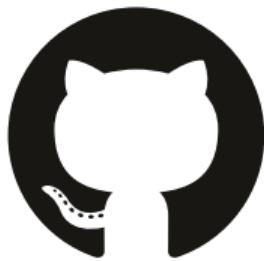
Testing different cooling-rate profiles in a box model



Testing different cooling-rate profiles in a box model



100%  python™ open-source code:

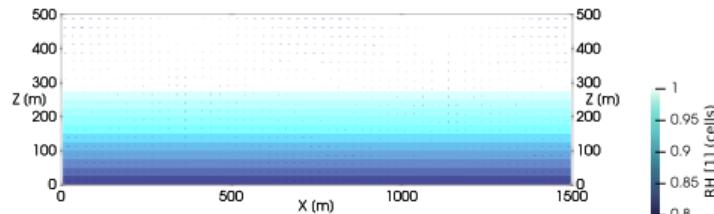


/ OPEN**ATMOS** / PySDM



Testing three flow regimes and two immersion freezing representations

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

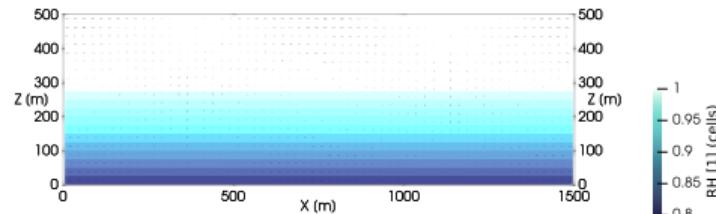


$w_{\max} \approx 1 \text{ m/s}$

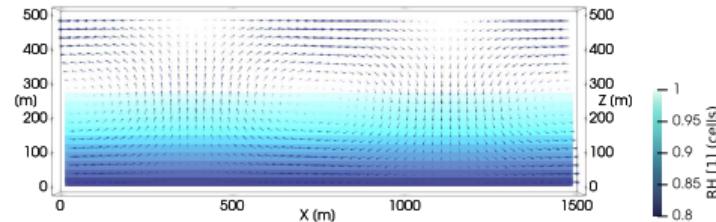
$w_{\max} \approx 3 \text{ m/s}$

Testing three flow regimes and two immersion freezing representations

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



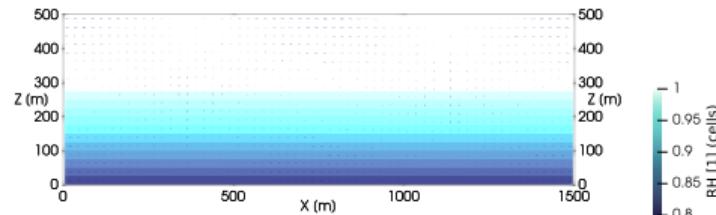
$w_{\max} \approx 1 \text{ m/s}$



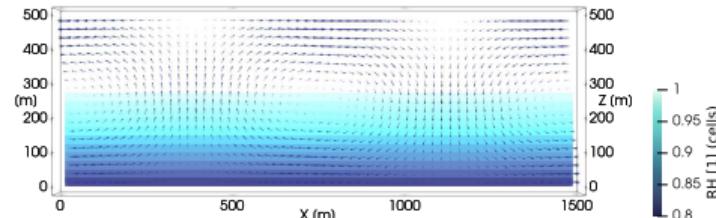
$w_{\max} \approx 3 \text{ m/s}$

Testing three flow regimes and two immersion freezing representations

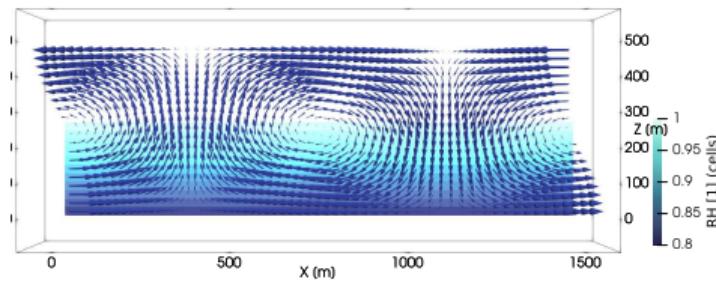
$w_{\max} \approx 1/3 \text{ m/s}$



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

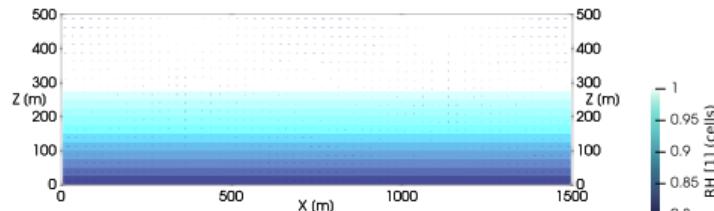


$w_{\max} \approx 3 \text{ m/s}$

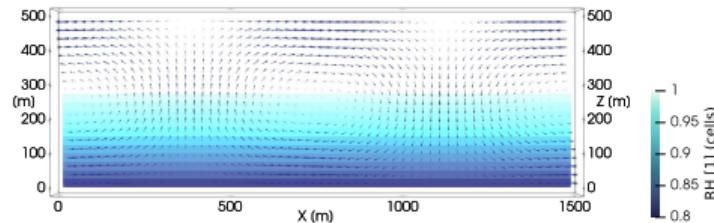


Testing three flow regimes and two immersion freezing representations

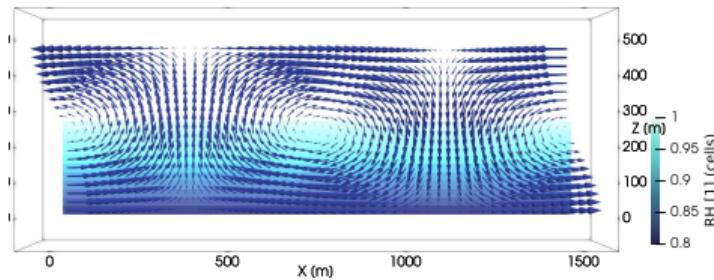
$w_{\max} \approx 1/3 \text{ m/s}$



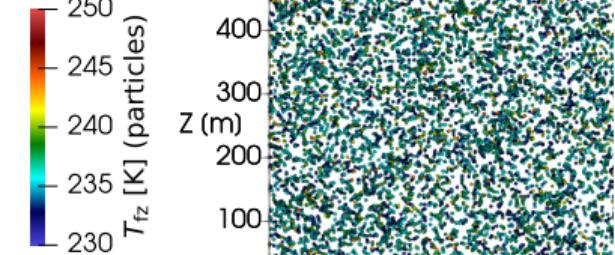
$w_{\max} \approx 1 \text{ m/s}$



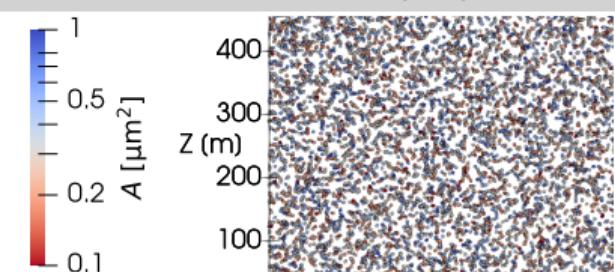
$w_{\max} \approx 3 \text{ m/s}$



singular (INAS)

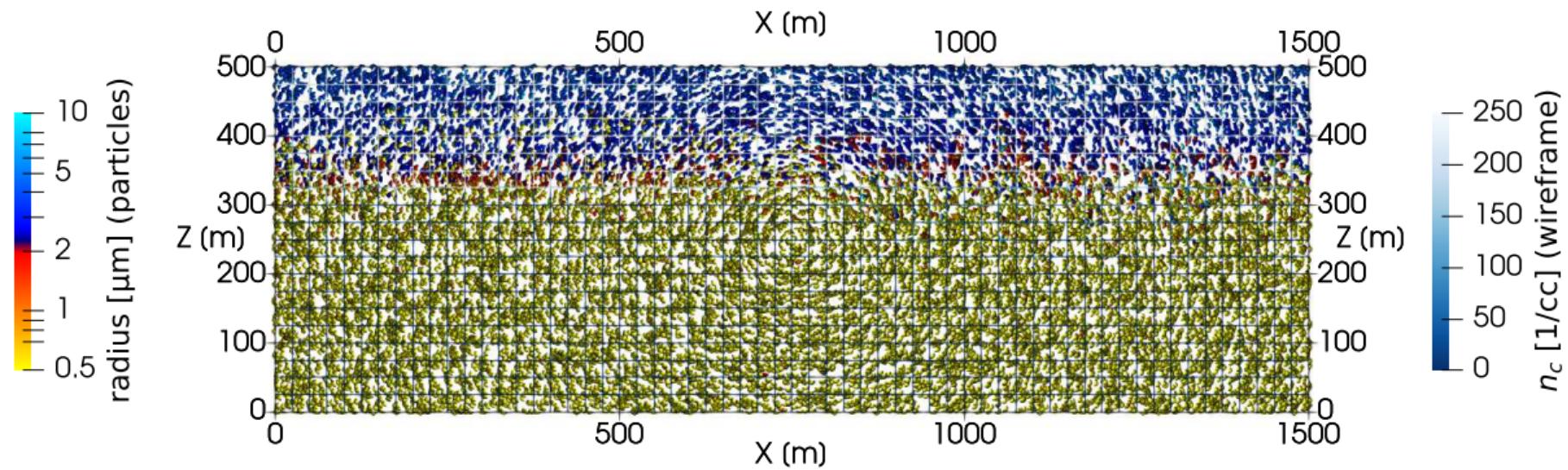


time-dependent (J_{het})



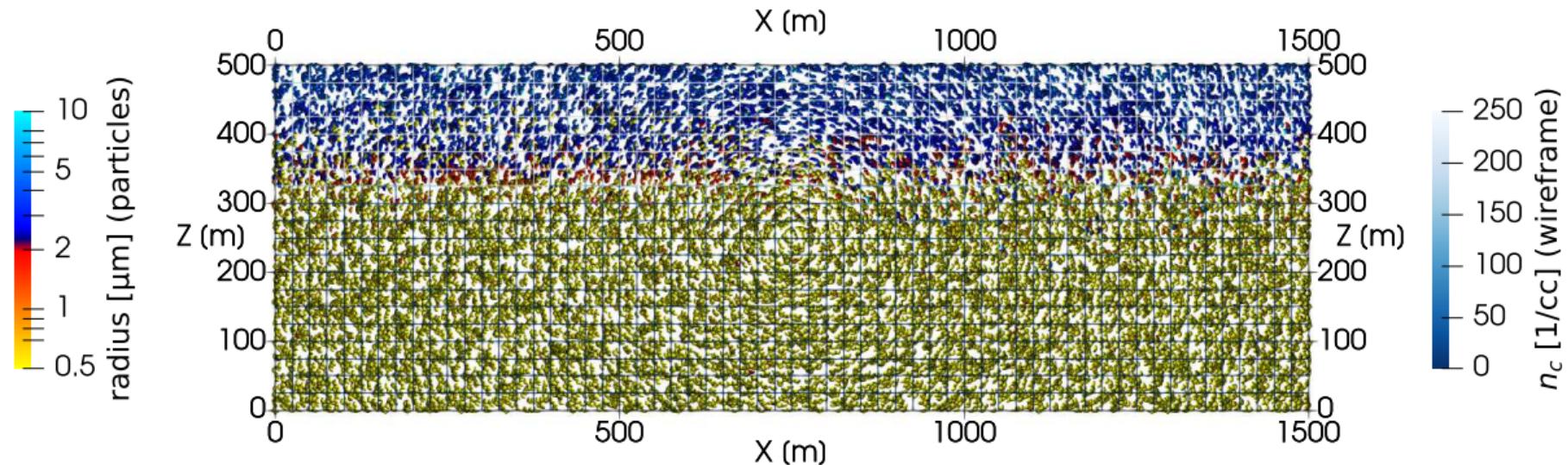
Particle-based μ -physics + prescribed-flow: spin-up

Time: 360 s (spin-up till 600.0 s)



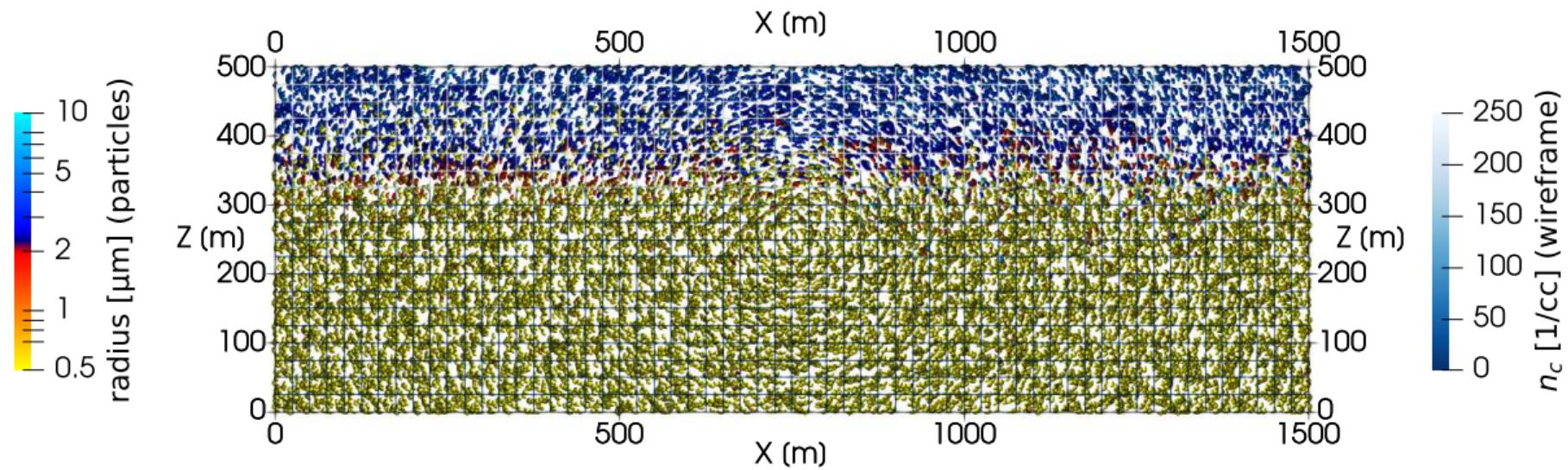
Particle-based μ -physics + prescribed-flow: spin-up

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



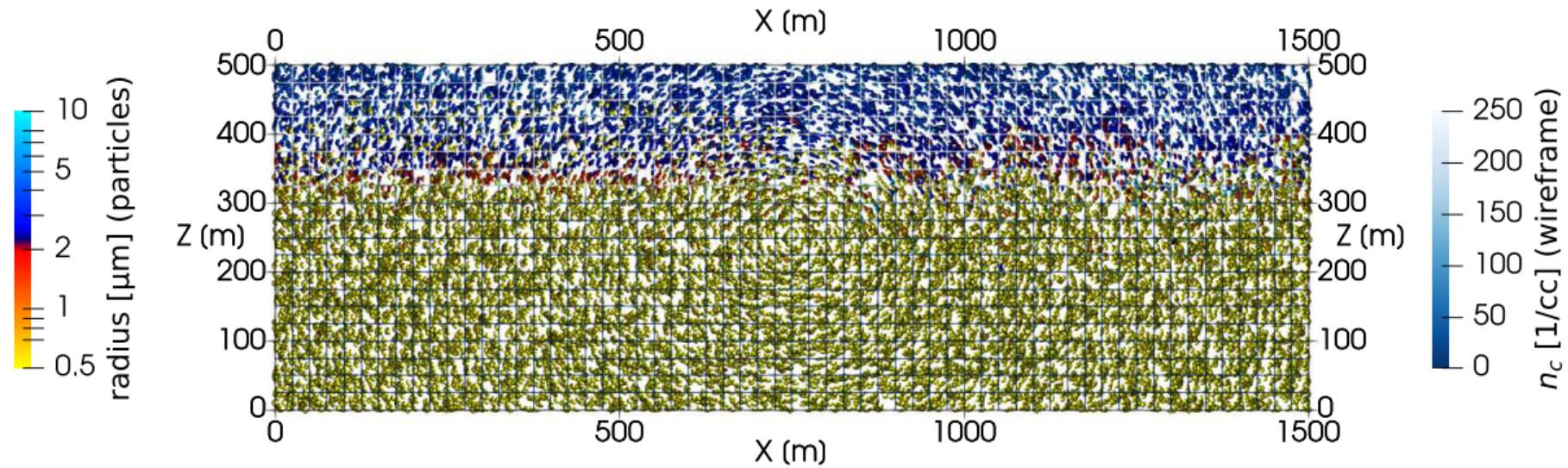
Particle-based μ -physics + prescribed-flow: spin-up

Time: 420 s (spin-up till 600.0 s)



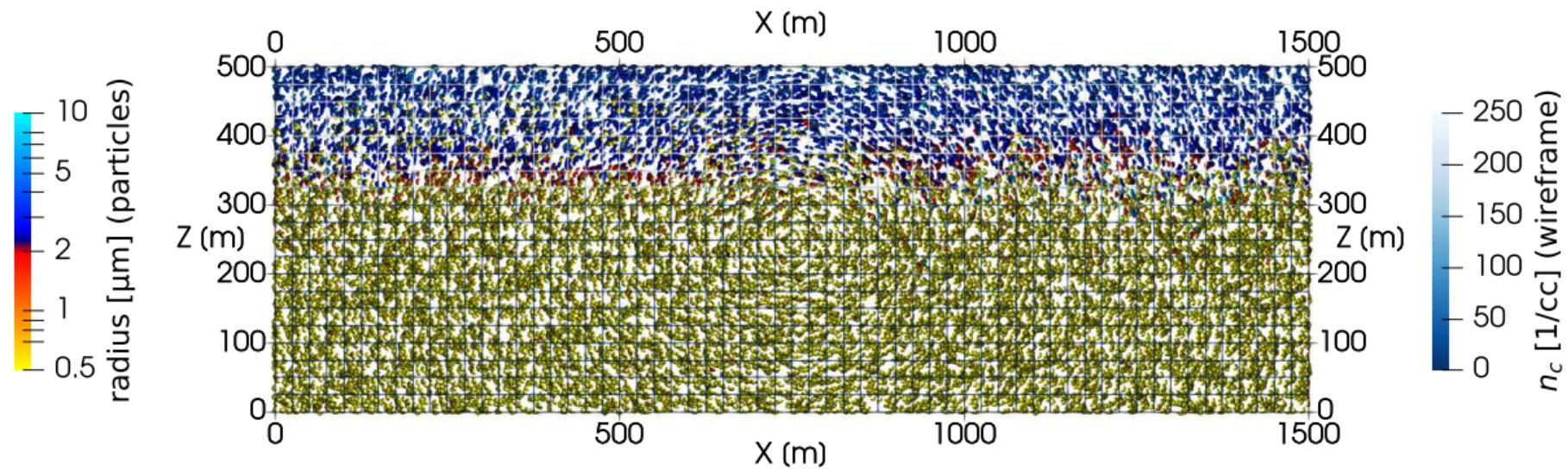
Particle-based μ -physics + prescribed-flow: spin-up

Time: 450 s (spin-up till 600.0 s)



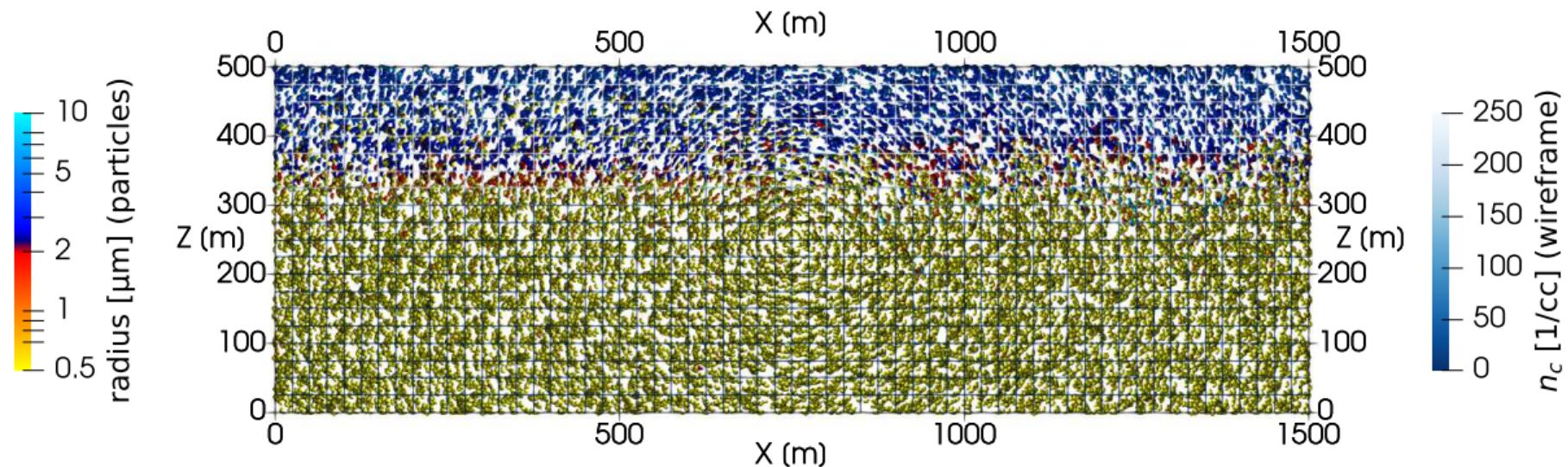
Particle-based μ -physics + prescribed-flow: spin-up

Time: 480 s (spin-up till 600.0 s)



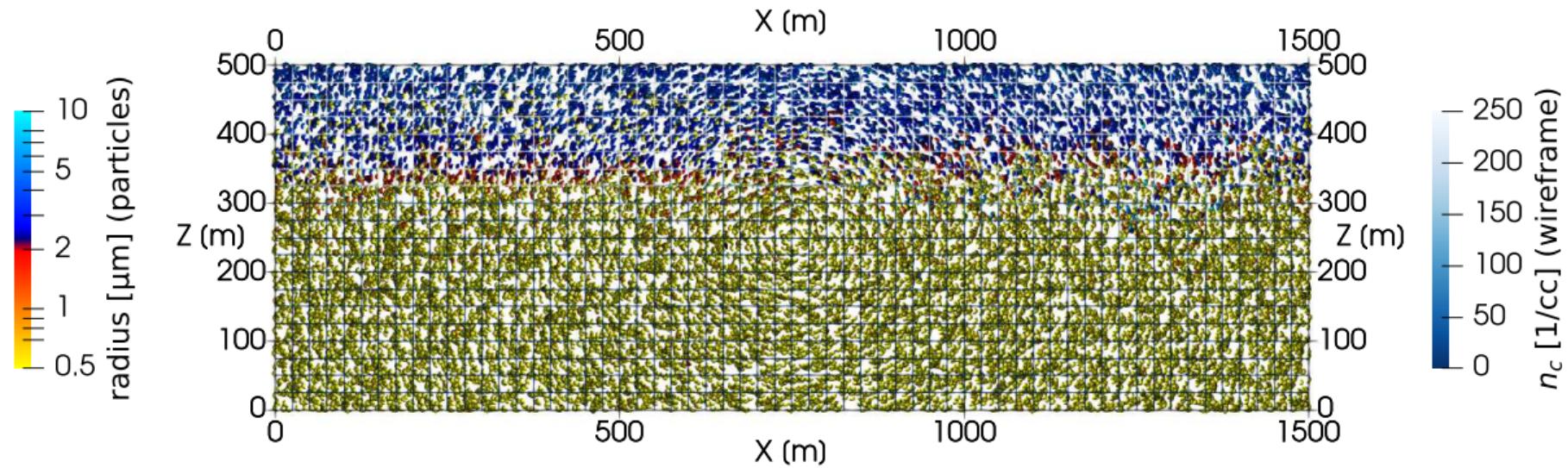
Particle-based μ -physics + prescribed-flow: spin-up

Time: 510 s (spin-up till 600.0 s)



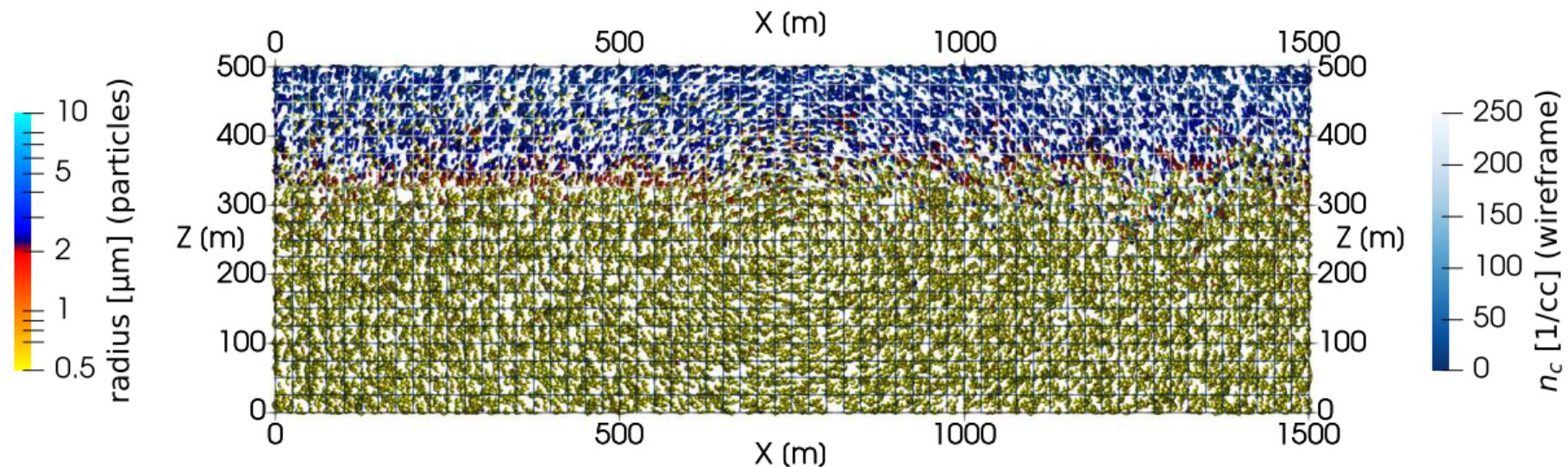
Particle-based μ -physics + prescribed-flow: spin-up

Time: 540 s (spin-up till 600.0 s)



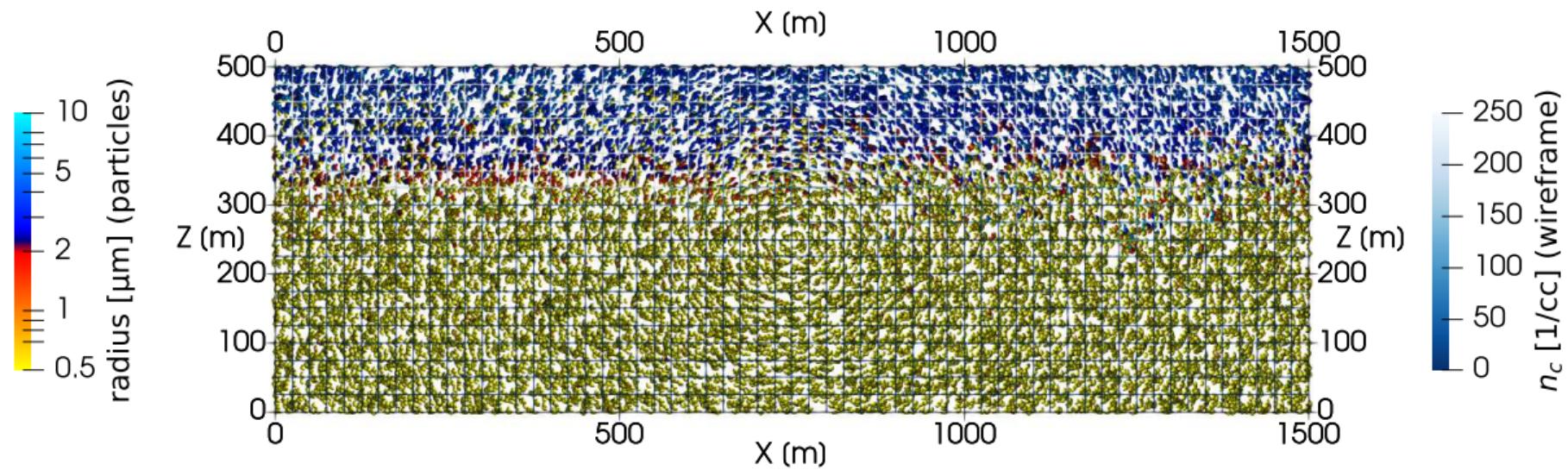
Particle-based μ -physics + prescribed-flow: spin-up

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



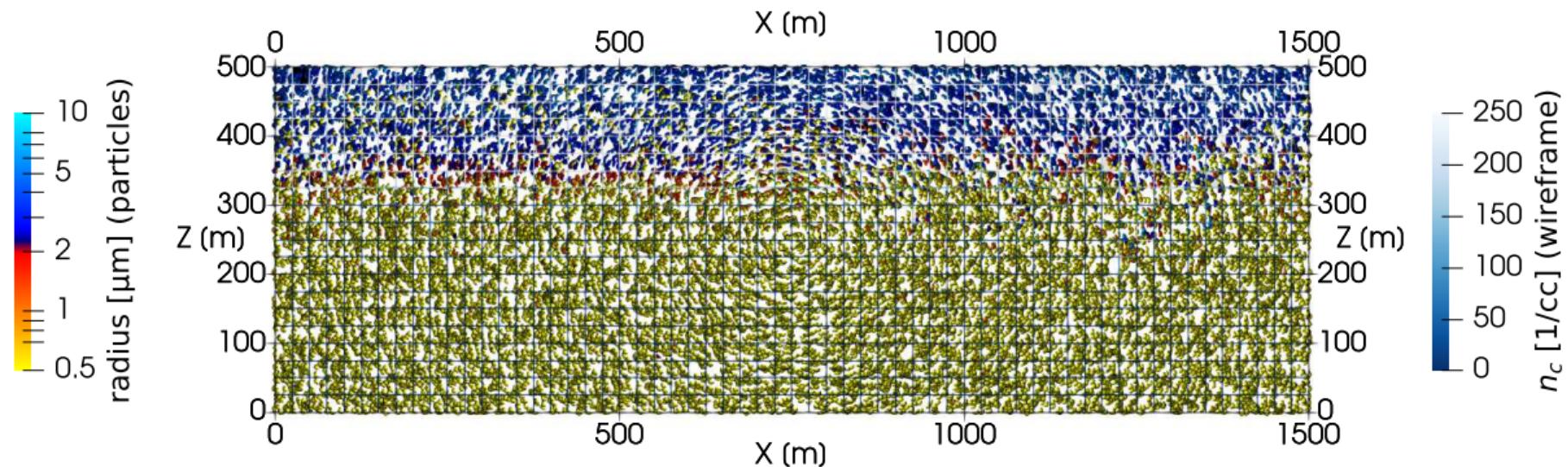
Particle-based μ -physics + prescribed-flow: spin-up

Time: 600 s (spin-up till 600.0 s)



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

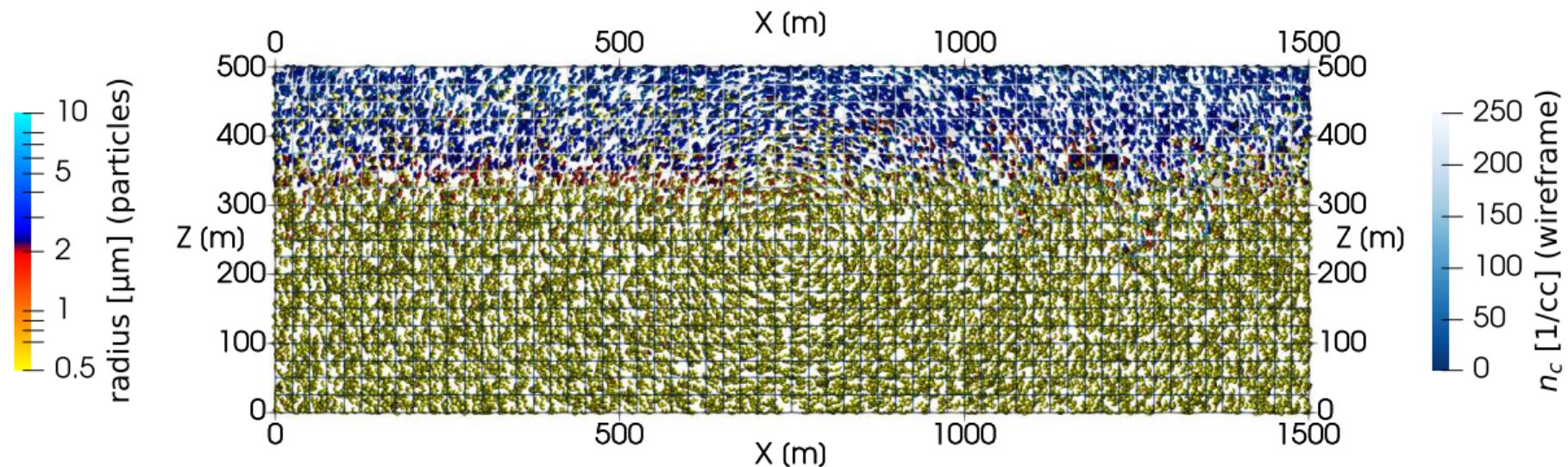
Time: 630 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \text{ }\mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

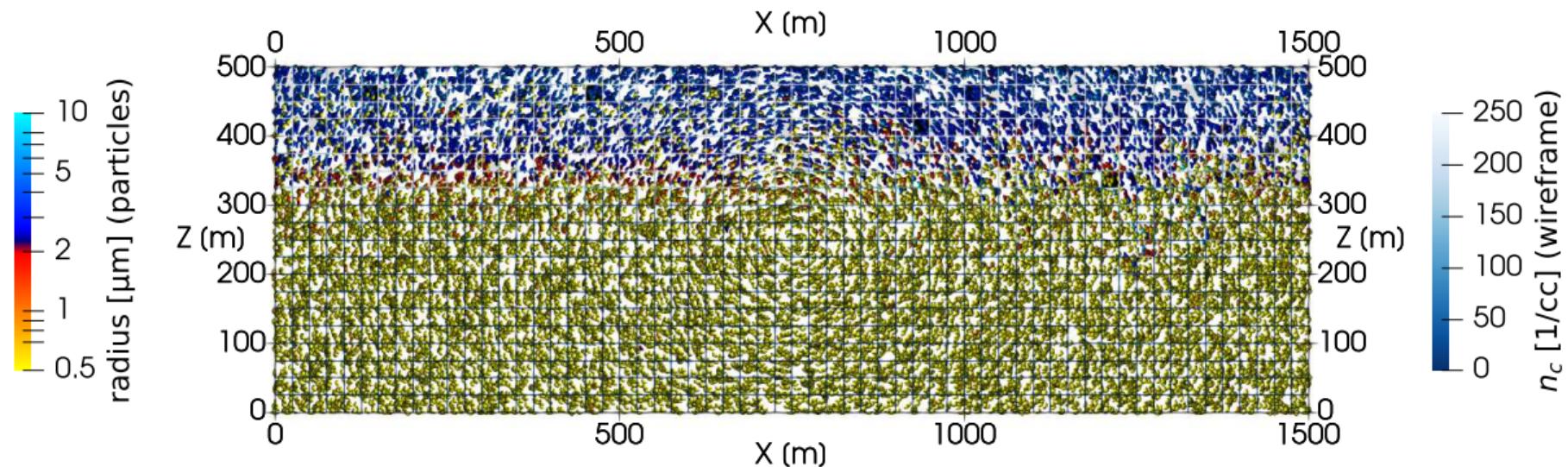
Time: 660 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
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Particle-based μ -physics + prescribed-flow: glaciation

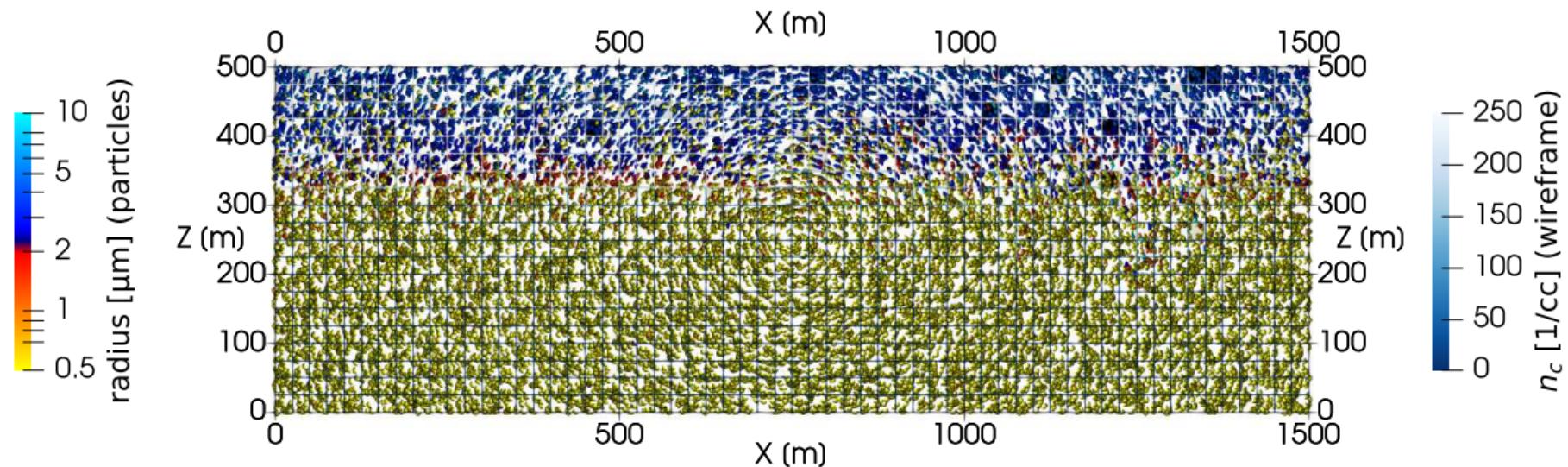
Time: 690 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
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Particle-based μ -physics + prescribed-flow: glaciation

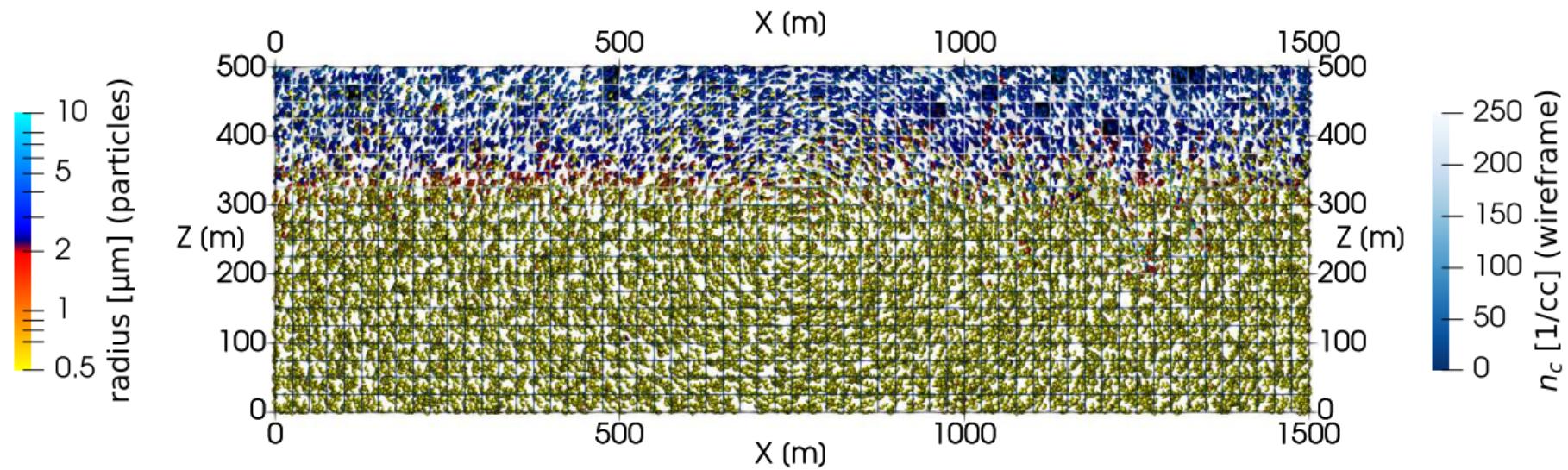
Time: 720 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
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Particle-based μ -physics + prescribed-flow: glaciation

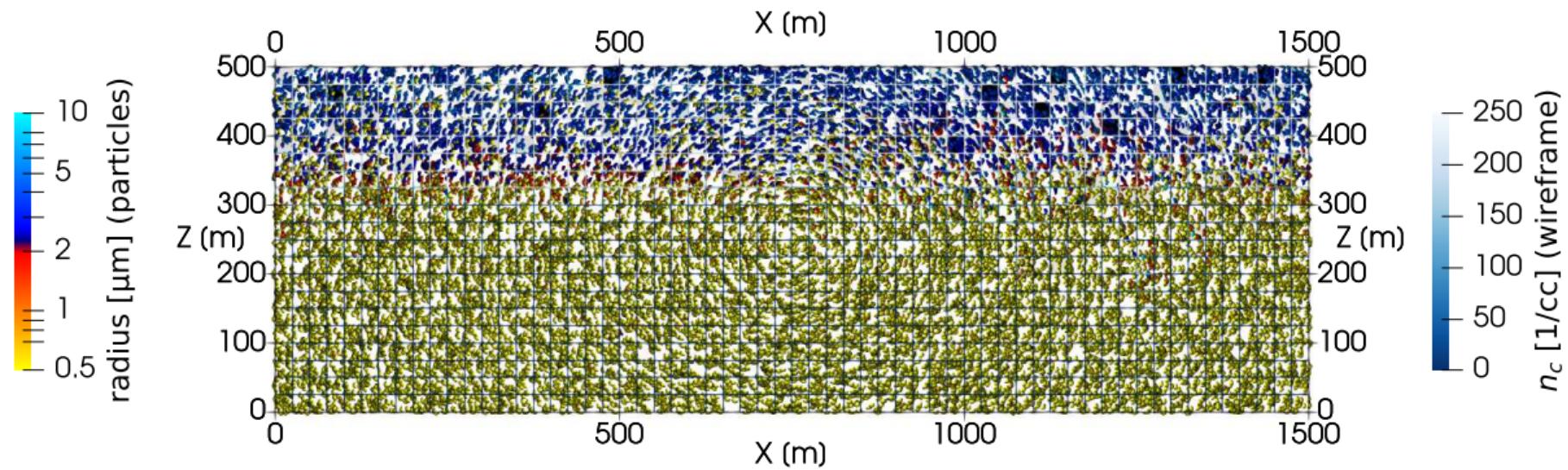
Time: 750 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
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Particle-based μ -physics + prescribed-flow: glaciation

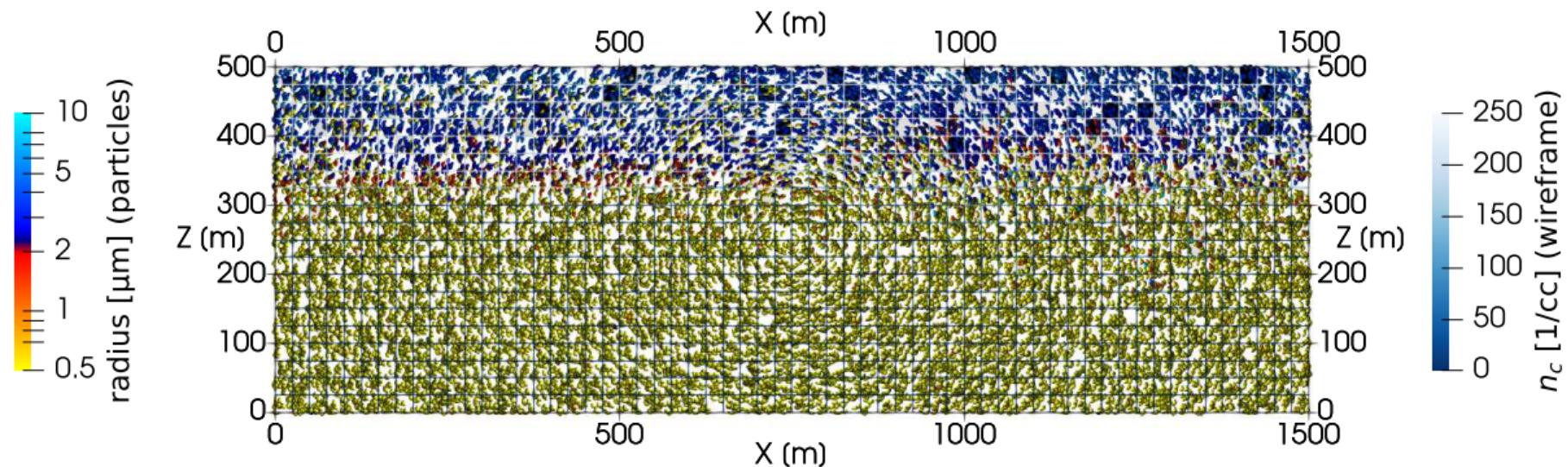
Time: 780 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \text{ } \mu\text{m}$, $\sigma_g = 2.55$)
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Particle-based μ -physics + prescribed-flow: glaciation

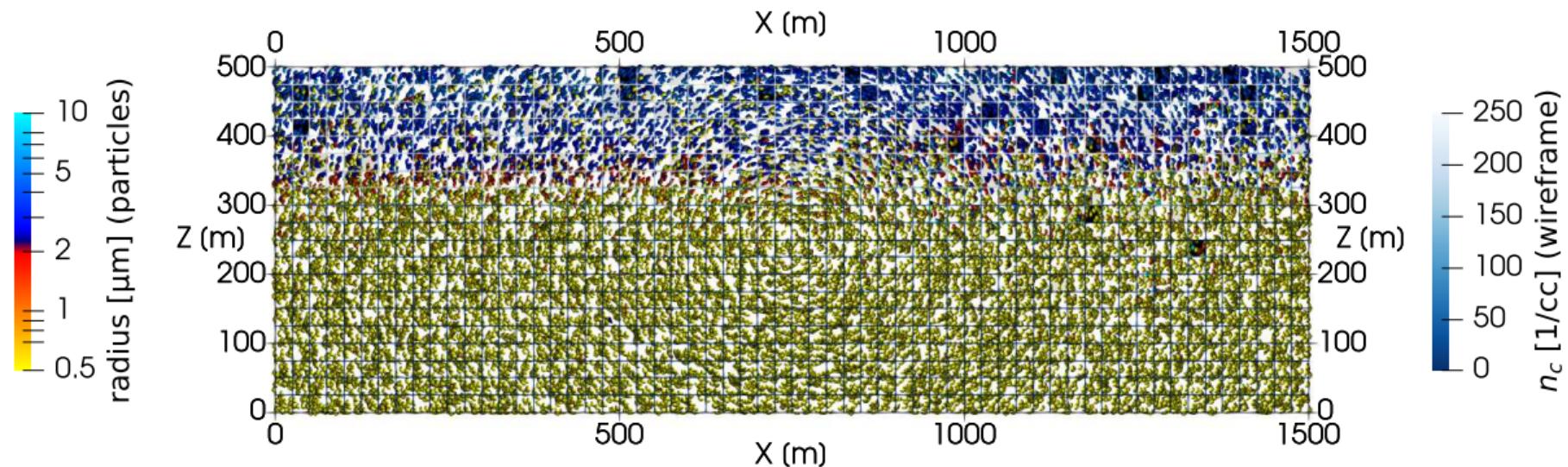
Time: 810 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
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Particle-based μ -physics + prescribed-flow: glaciation

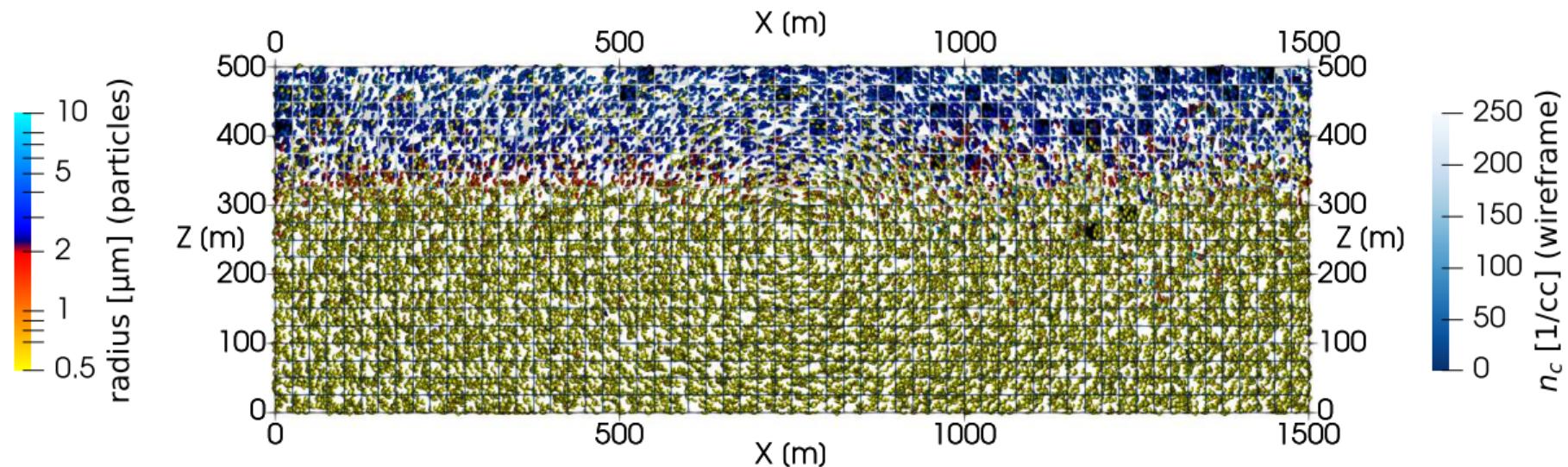
Time: 840 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \text{ } \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

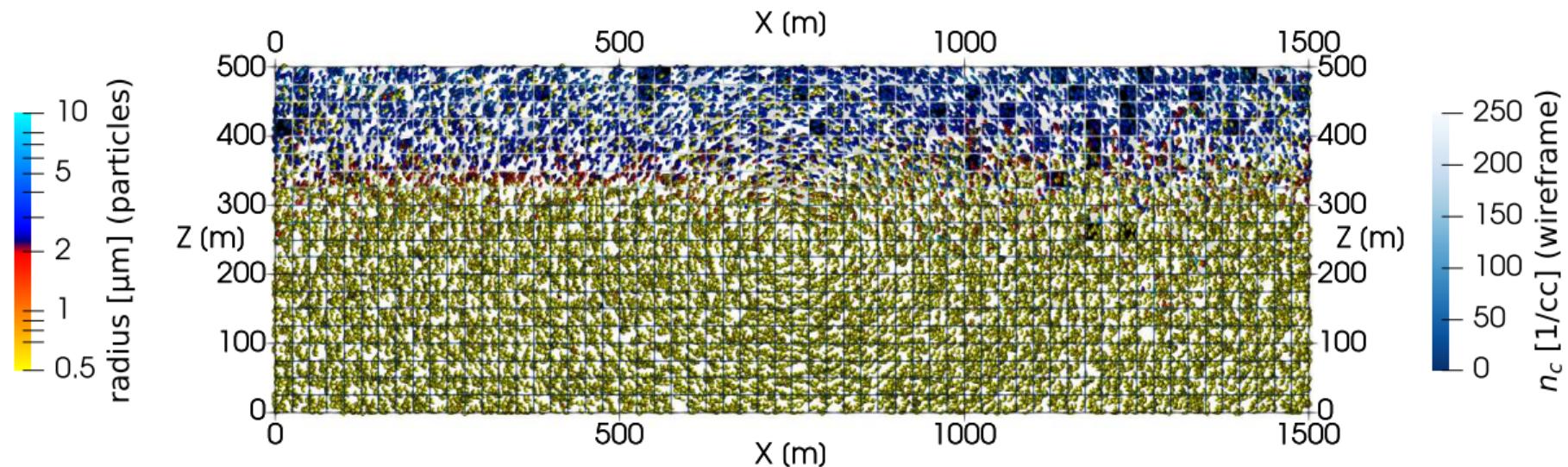
Time: 870 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \text{ } \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

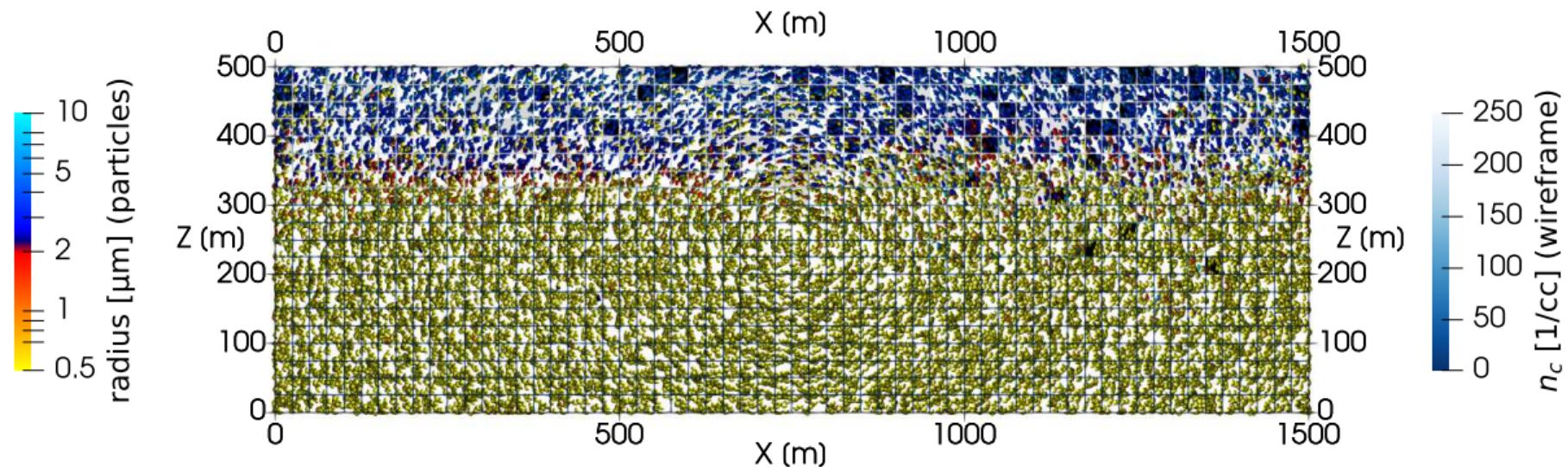
Time: 900 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \text{ μm}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

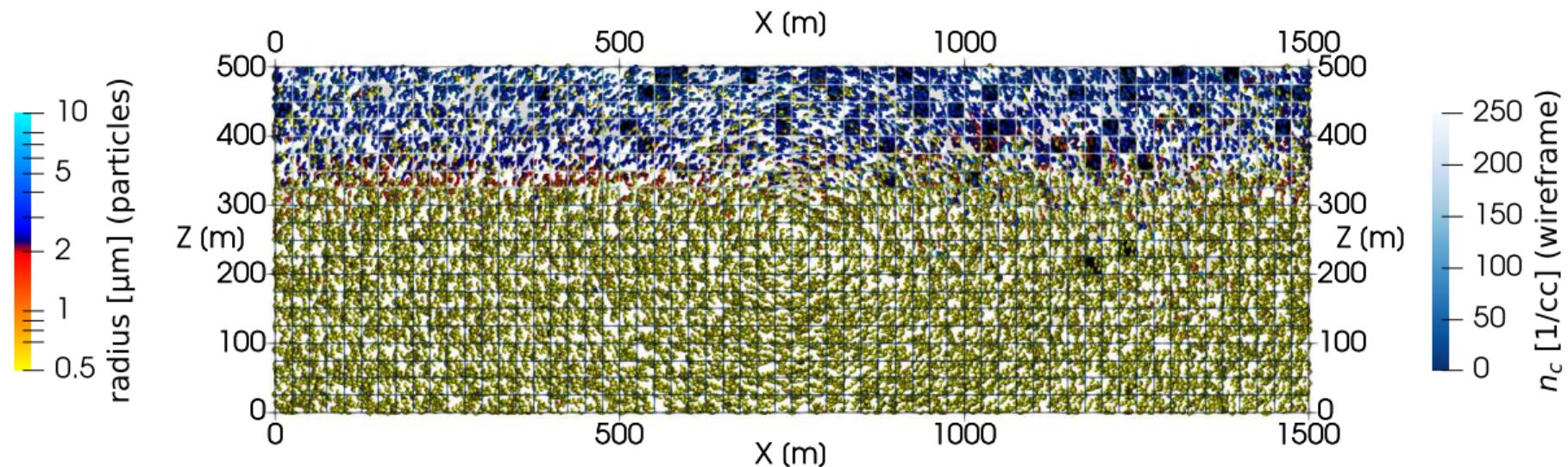
Time: 930 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

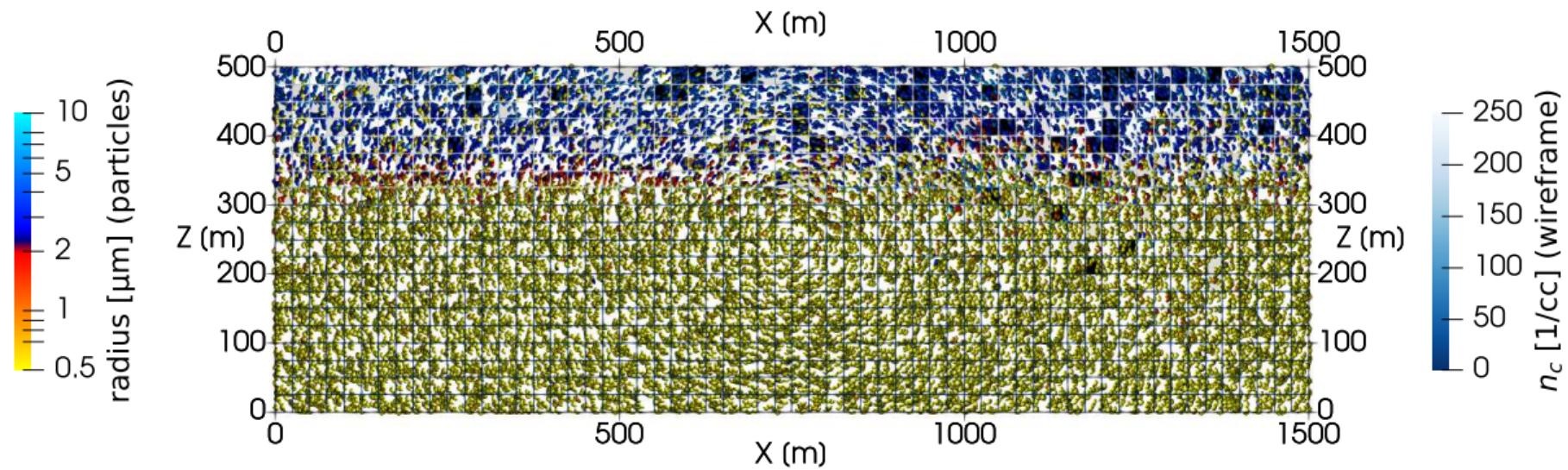
Time: 960 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

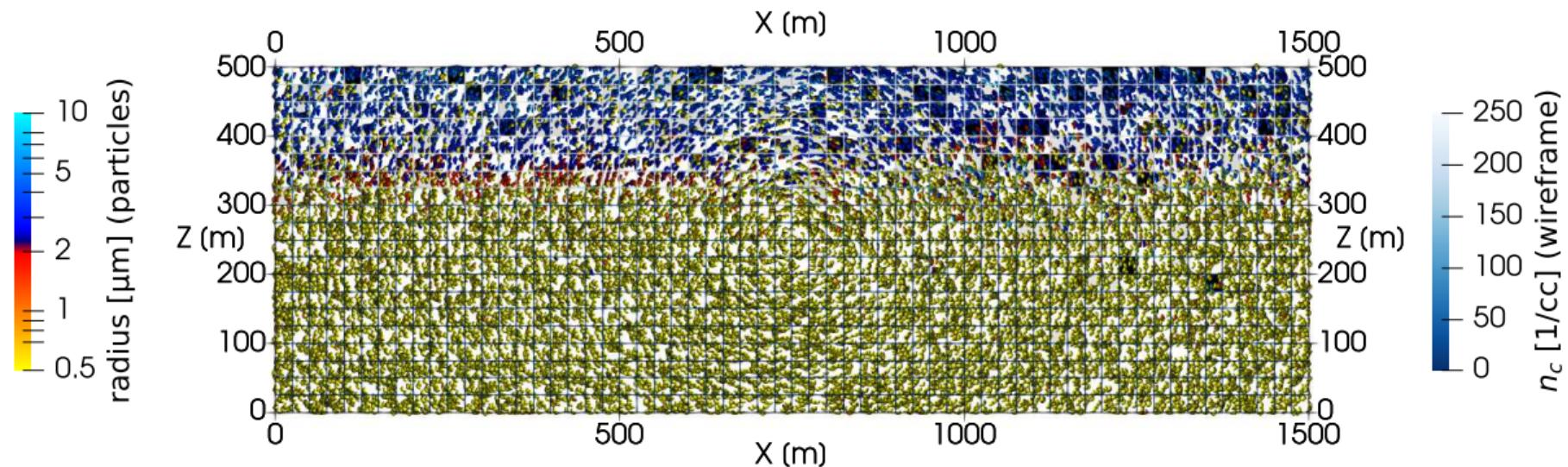
Time: 990 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

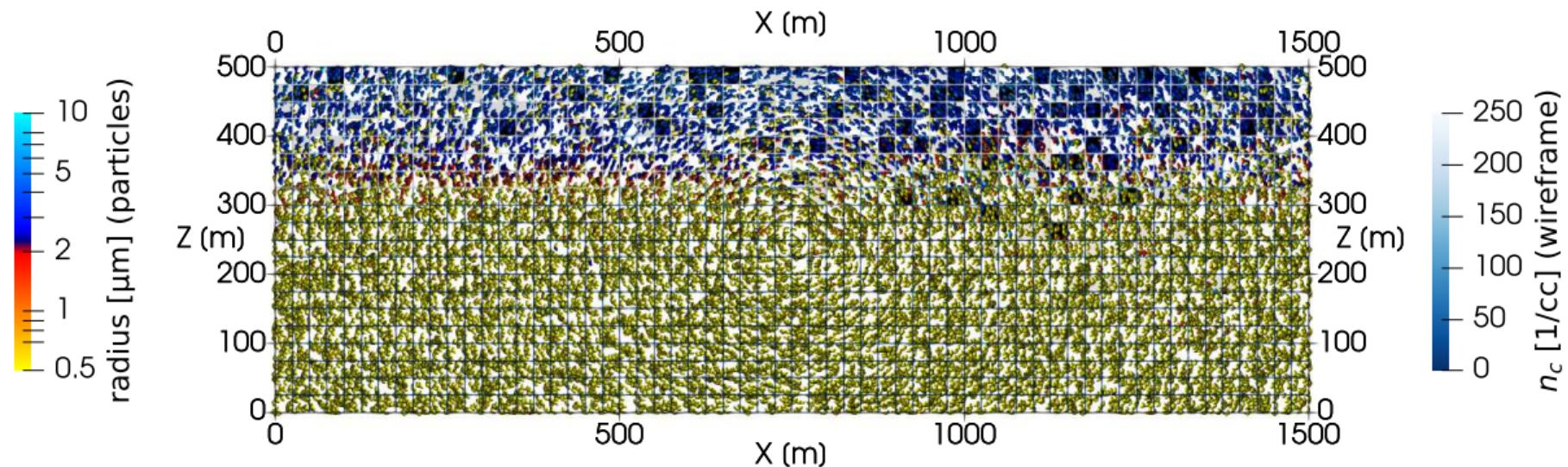
Time: 1020 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

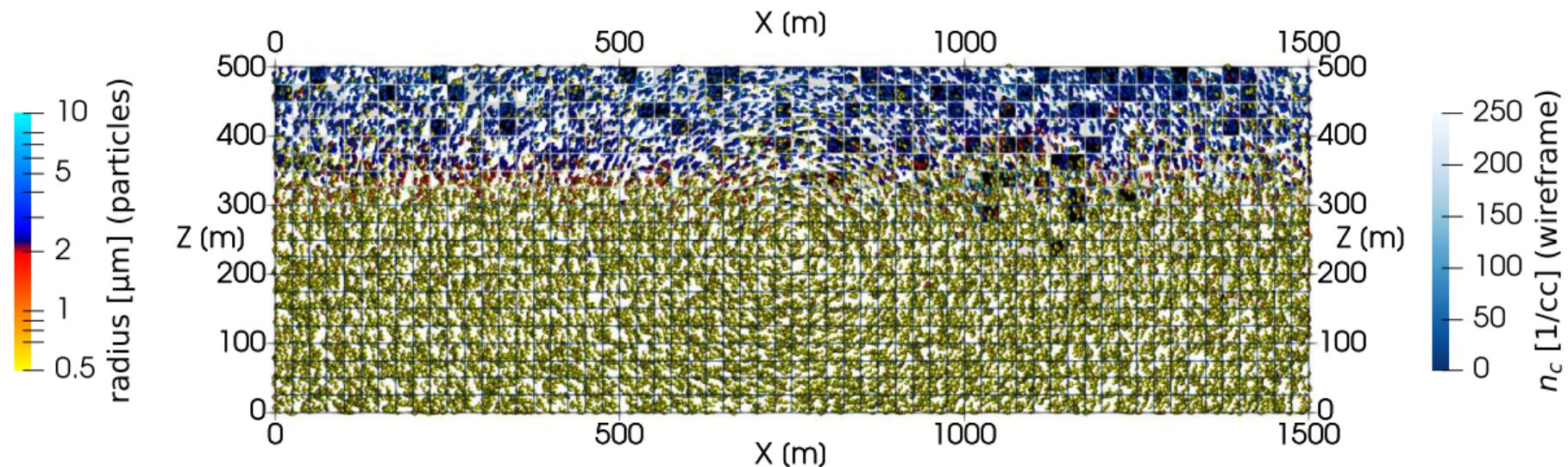
Time: 1050 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \text{ } \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

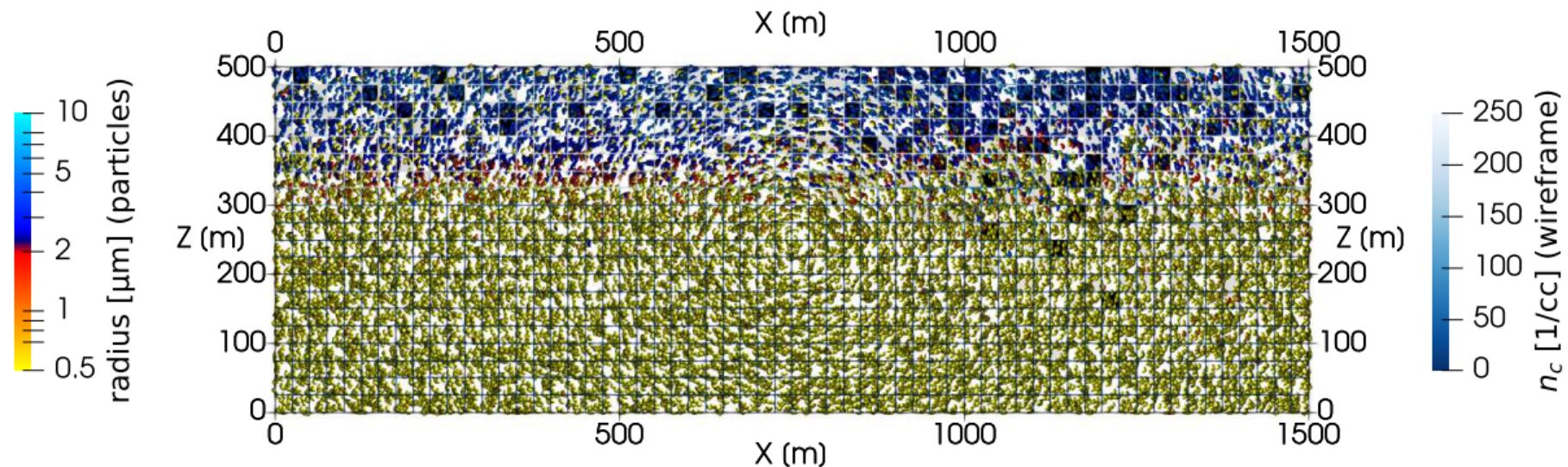
Time: 1080 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

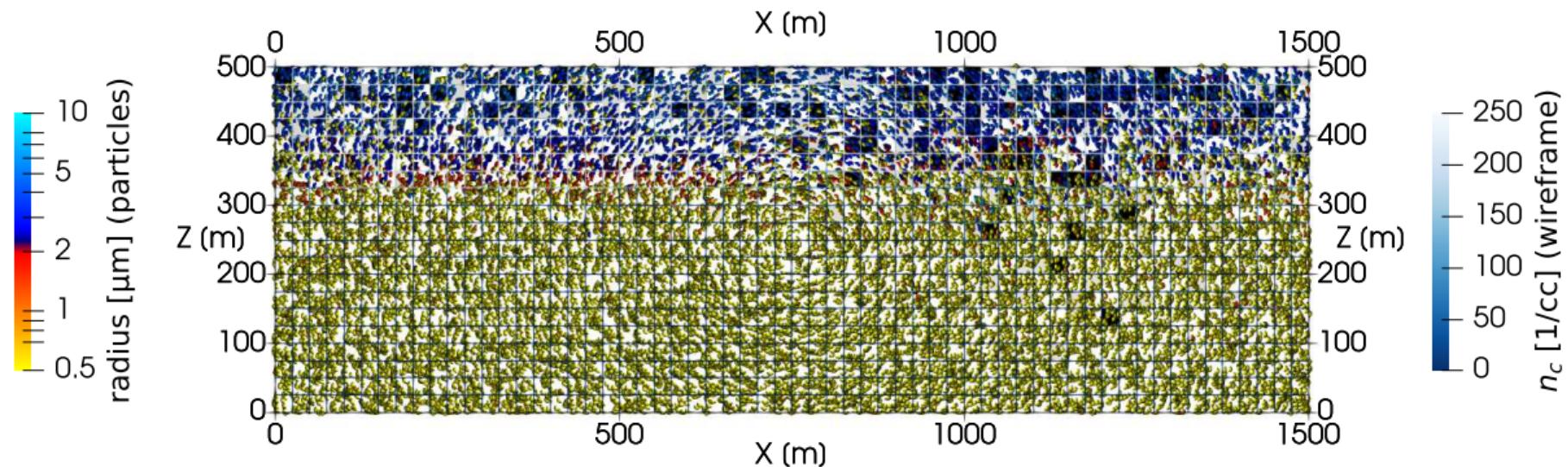
Time: 1110 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

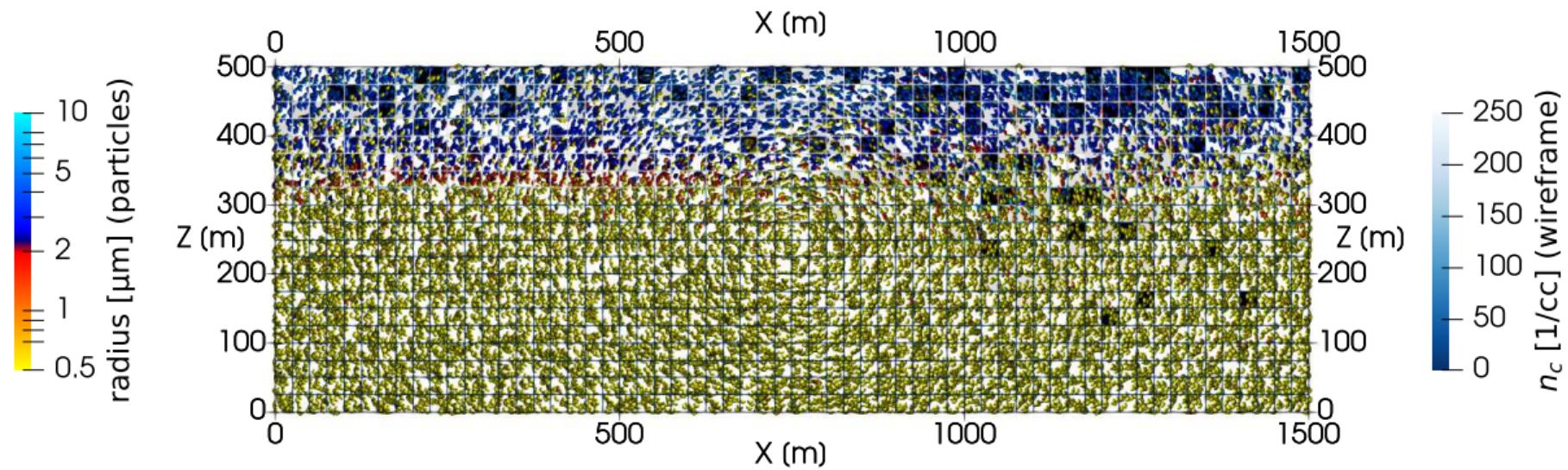
Time: 1140 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

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

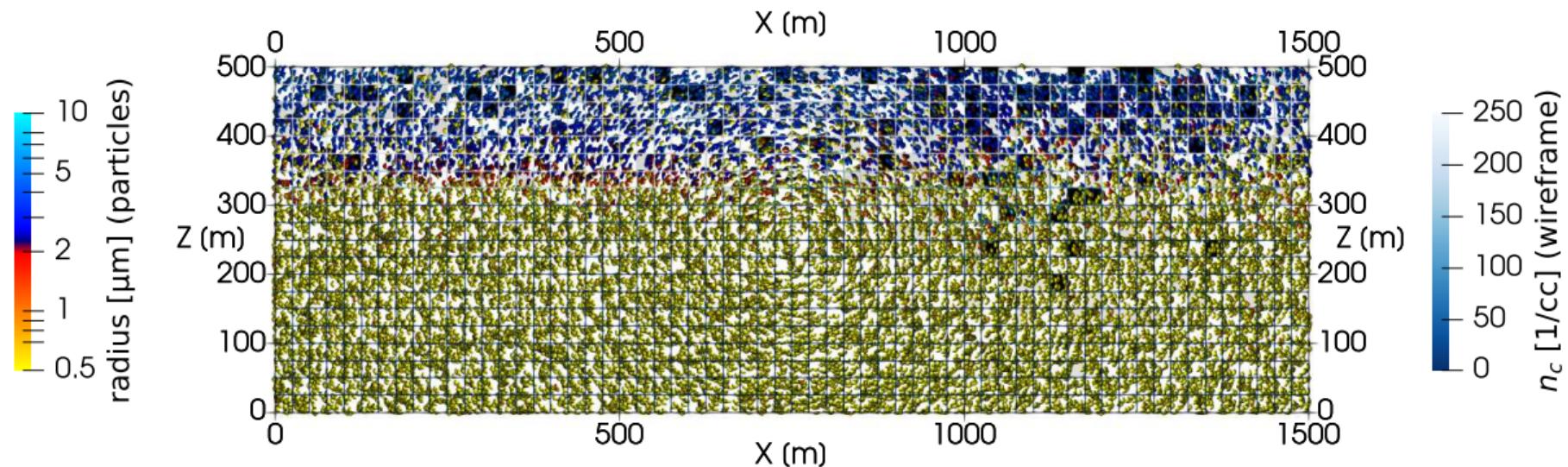
Time: 1170 s (spin-up till 600.0 s)



16+16 super-particles/cell for INP-rich + INP-free particles
 $N_{\text{aer}} = 300/\text{cc}$ (two-mode lognormal) $N_{\text{INP}} = 150/L$ (lognormal, $D_g = 0.74 \text{ } \mu\text{m}$, $\sigma_g = 2.55$)
spin-up = freezing off; subsequently frozen particles act as tracers

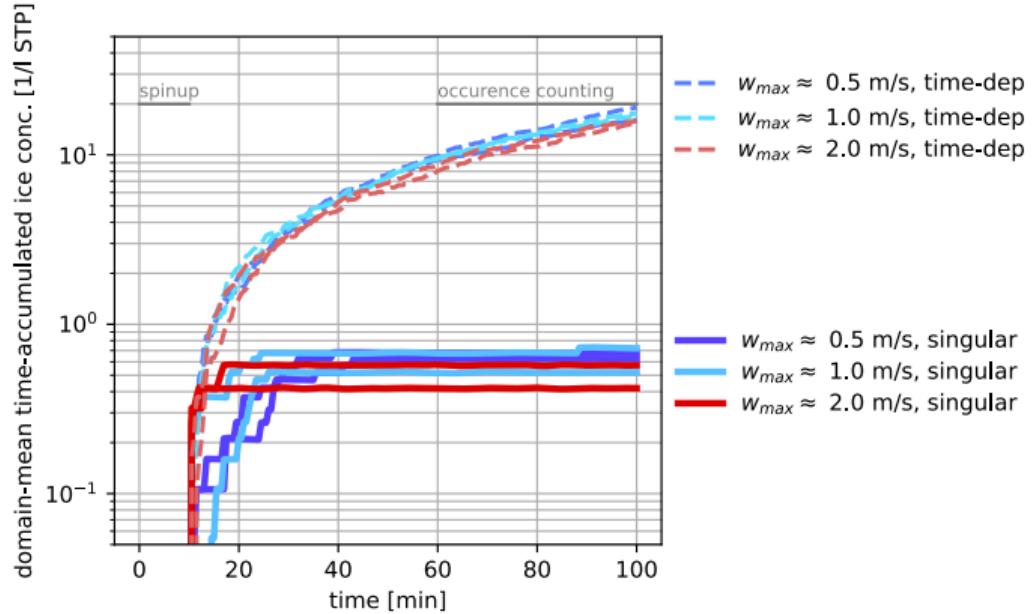
Particle-based μ -physics + prescribed-flow: glaciation

Time: 1200 s (spin-up till 600.0 s)

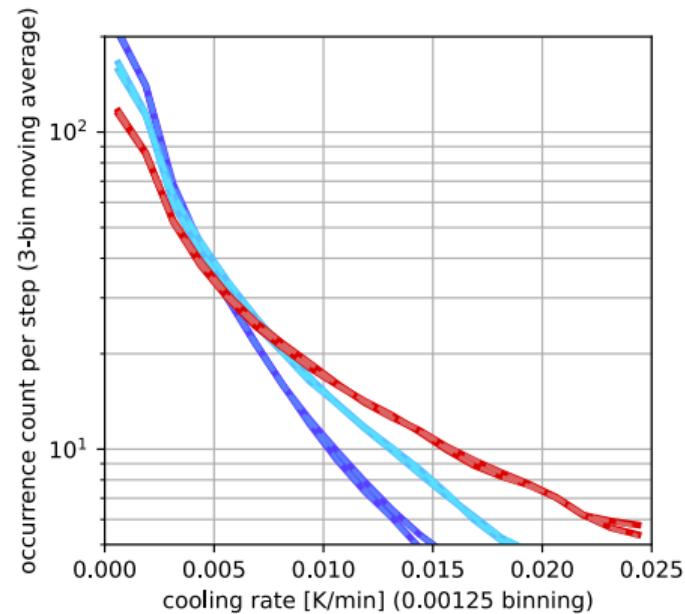
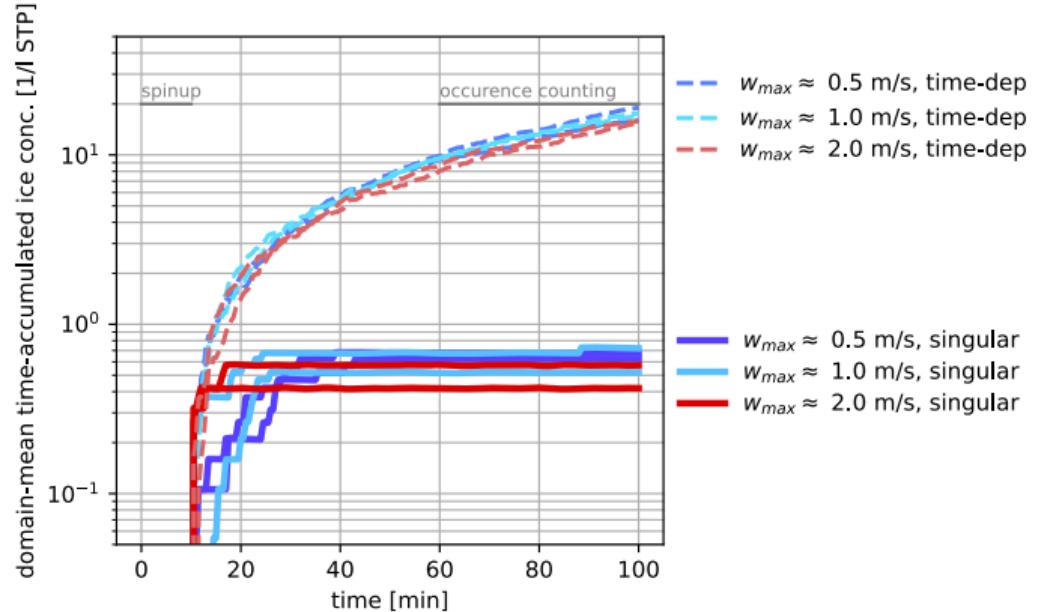


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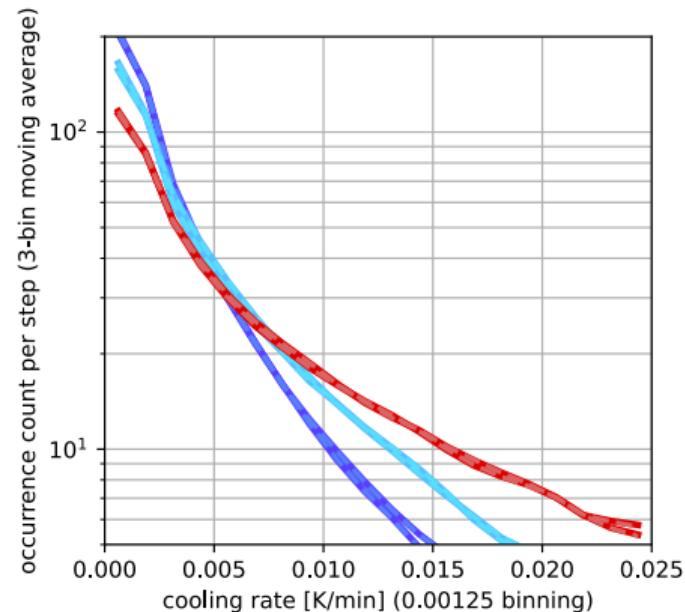
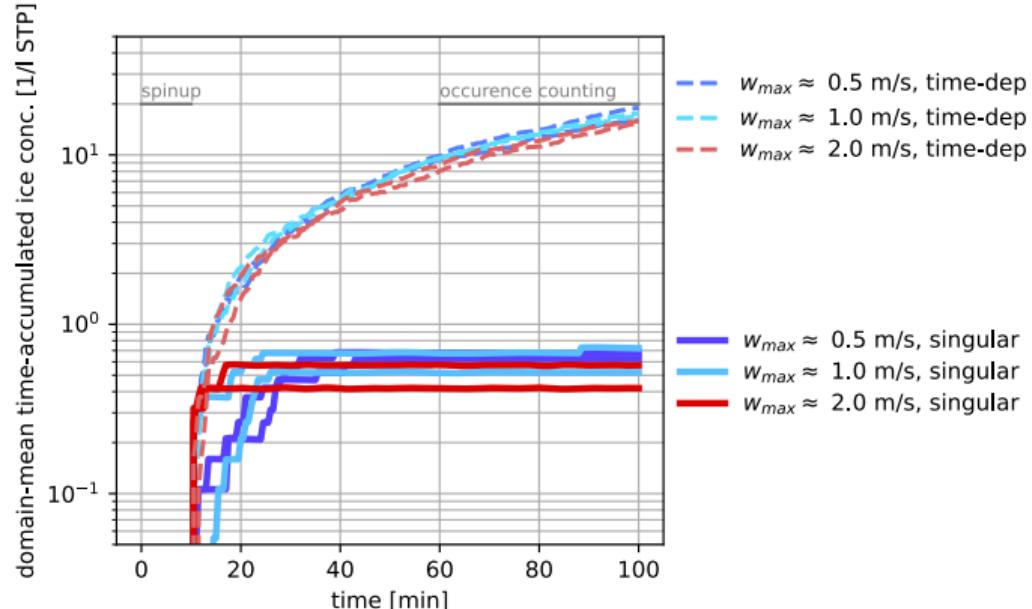
Testing three flow regimes and two immersion freezing representations



Testing three flow regimes and two immersion freezing representations

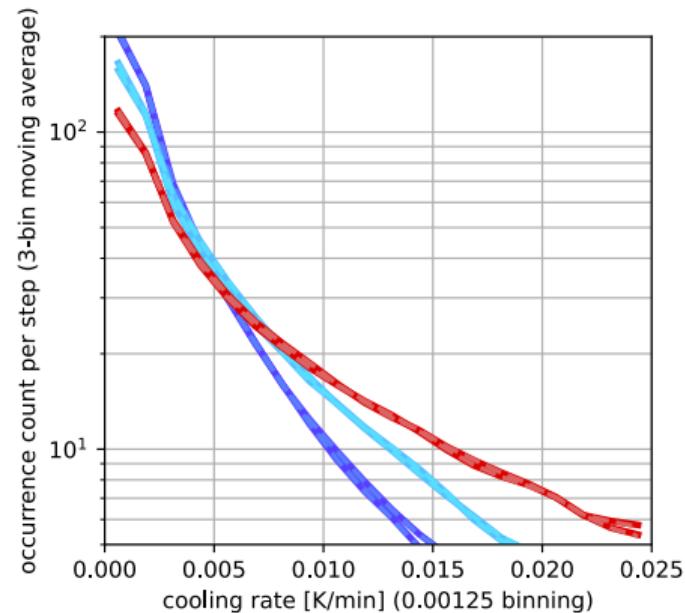
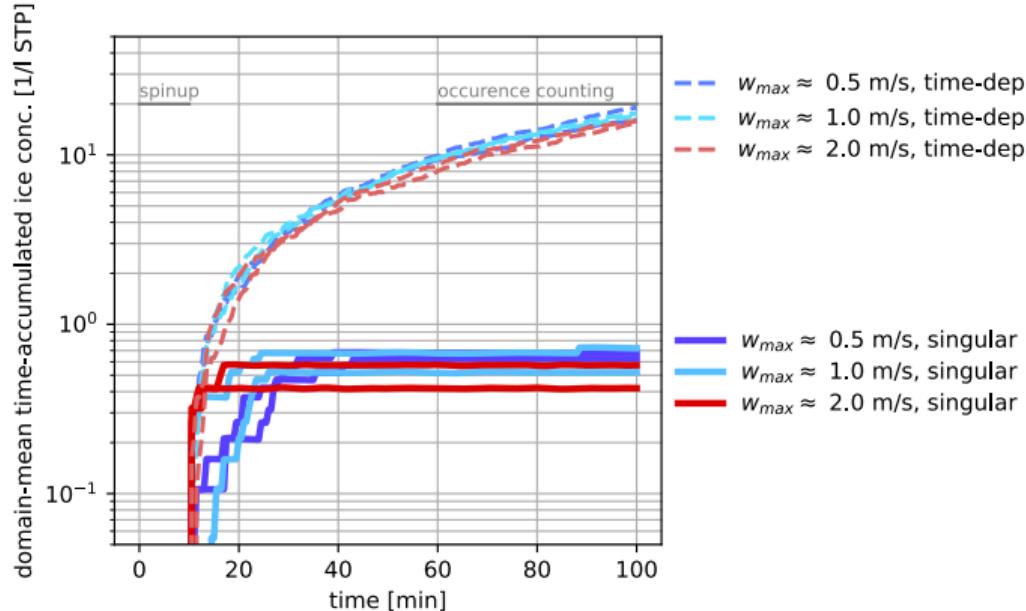


Testing three flow regimes and two immersion freezing representations



- range of cooling rates in simple flow (far from 0.5 K/min for AIDA as in Niemand et al. 2012)

Testing three flow regimes and two immersion freezing representations



- ▶ 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)

聞いてくれておおきに!

Thank you for your attention!

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Draft:Super Droplet Method

Contents

(Top)

SDM in the cloud microphysics model taxonomy

▼ SDM Monte-Carlo
Algorithm for
Coagulation of
Particles

Super-particle state

Well-mixed control volume

Attribute sampling

Time stepping.

In mathematical modeling of aerosols, clouds and precipitation, **Super Droplet Method (SDM)** is a Monte-Carlo approach for representing collisions and coalescence of particles in atmospheric fluid dynamics simulations. The method and its name was introduced in a 2007 arXiv e-print by Shin-ichiro Shima et al.^[1] (preceded by a 2006 patent application^[2] and followed by 2008 RIMS Kôkyûroku^[3] and 2009 QJRMS^[4] papers).

SDM algorithm is a probabilistic alternative to the deterministic model of the process embodied in the [Smoluchowski coagulation equations](#). Among the key characteristics of SDM is that it is not subject to the "[curse of dimensionality](#)" that hampers application of other methods when multiple particle attributes need to be resolved in a simulation^[7]. The algorithm is [embarrassingly parallel](#), has [linear time complexity](#), constant state vector size (number conservation of simulated particles during collisions)

