

# **Impact of nucleation rates and diffusional growth on ice nucleation events**

**EGU2020: Sharing Geoscience Online**

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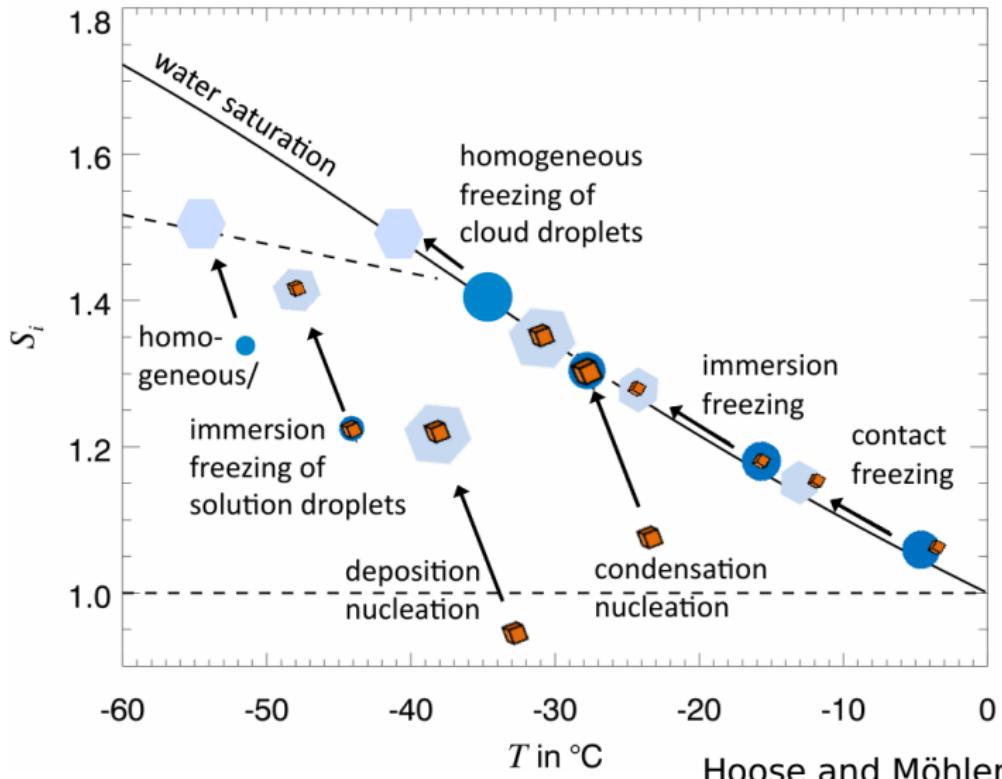
## Ice nucleation

- ▶ Ice nucleation is important process for formation and evolution of clouds containing ice particles
- ▶ Ice nucleation is not well understood as a process (even on molecular scale)
- ▶ Formulation of ice nucleation is uncertain
- ▶ Formulation of ice nucleation in models is often not consistent

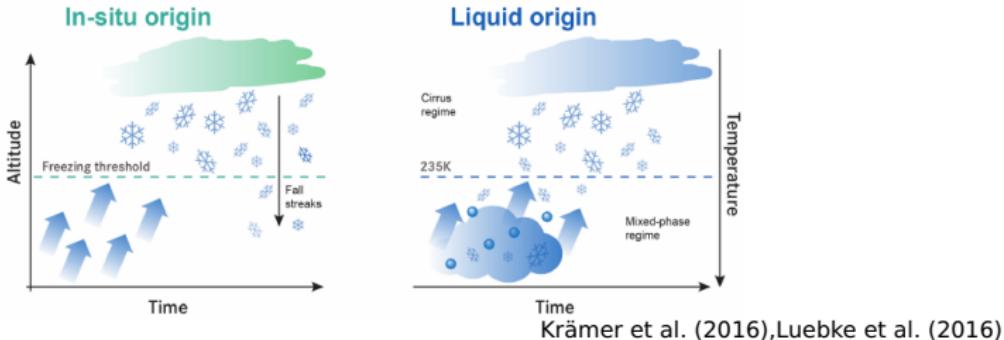
## Consequences

- ▶ Impact of ice nucleation on evolution of clouds uncertain
- ▶ Relevance of correct formulation for evolution of clouds in coarse models not clear

# Nucleation pathways



Hoose and Möhler (2012)



- ▶ **Liquid origin: pre-existing cloud droplets**

$$S \sim 1, S_i > 1, \sim 235 \text{ K} \leq T \leq 273 \text{ K}$$

- ▶ without solid aerosol particles: homogeneous freezing of (large) pure water or solution droplets
- ▶ with solid aerosol particles: immersion/contact freezing

- ▶ **In situ origin: no pre-existing water available**

$$S < 1, S_i > 1 \text{ and } T < 235 \text{ K}$$

- ▶ without solid aerosol particles: homogeneous freezing of (small) solution droplets (i.e. haze)
- ▶ with solid aerosol particles: deposition freezing

## Process

- ▶ controlled laboratory experiments (bulk)
- ▶ aqueous solution droplets freeze stochastically
- ▶ anorganic solute does not play a role

## Formulation

$$J = J_0 \exp(P_3(\Delta a_w)), \quad \Delta a_w = a_w - a_w^i \quad (1)$$

with  $P_3$  polynomial of degree 3, see Koop et al. (2000)

## Questions

- ▶ How sensitive are nucleation events to the exact formulation of the nucleation rate?
- ▶ Can other processes also affect nucleation events in a critical way?

## Outline

- ▶ Approximations to the original formulation of nucleation rate
- ▶ Idealized simulations of nucleation events
- ▶ Investigations of the sensitivity of nucleation events due to changes in the representation

## Boxmodel approach

- ▶ Constant temperature ( $T = 196 / 216 / 236 \text{ K}$ ) and pressure ( $p = 200 \text{ hPa}$ )
- ▶ Cooling rate/vertical velocity:  $0.01 \text{ ms}^{-1} \leq w \leq 10 \text{ ms}^{-1}$
- ▶ Increase of supersaturation  $S_i$
- ▶ Processes: Nucleation and diffusional growth
- ▶ Background aerosol: sulphuric acid,  $N_a \sim 10^5 \text{ cm}^{-3}$
- ▶ Integration with very small time step  $\Delta t = 0.01 \text{ s}$

Comparison to former studies (Kärcher and Lohmann, 2002;  
Spichtinger and Gierens, 2009)

## Thermodynamics

$$a_w = \frac{p_{\text{sol}}}{p_{\text{liq}}} \stackrel{\text{in eq.}}{=} \frac{p_v}{p_{\text{liq}}} = S_{\text{liq}} \quad (2)$$

$$\Delta a_w = a_w - a_w^i = \frac{p_v}{p_{\text{liq}}(T)} - \frac{p_{\text{ice}}(T)}{p_{\text{liq}}(T)} = (S_i - 1) \frac{p_{\text{ice}}(T)}{p_{\text{liq}}(T)} = (S_i - 1) a_w^i(T) \quad (3)$$

Two approximations

- ▶ Linear fit:

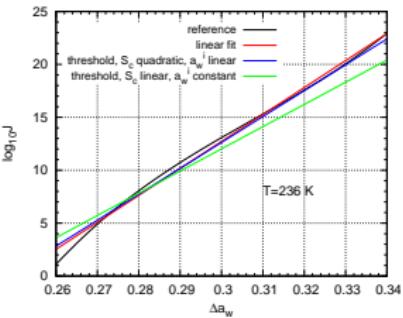
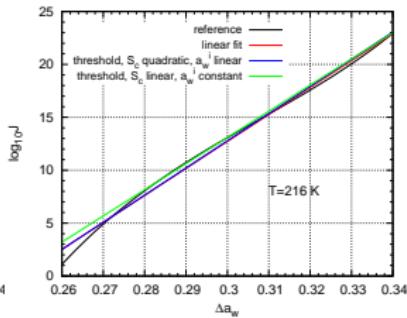
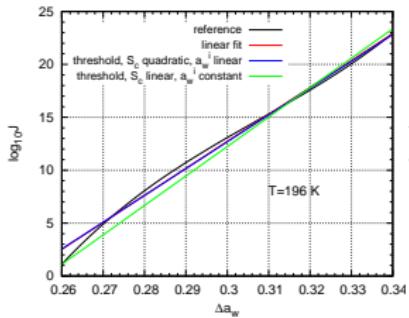
$$J \sim \exp(a_0 + a_1 \Delta a_w) \quad (4)$$

- ▶ Threshold description

$$J \sim \exp(A(T)(S_i - S_c(T))) \quad (5)$$

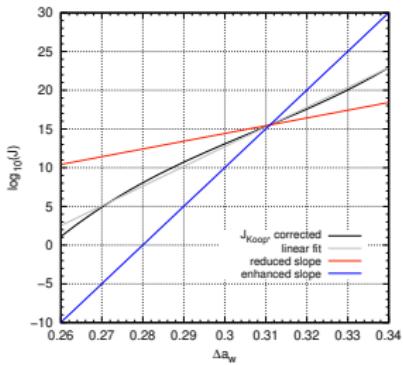
# Nucleation rates

## Approximations

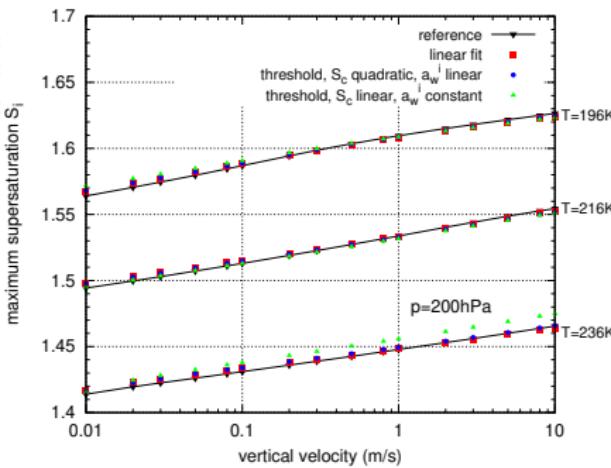
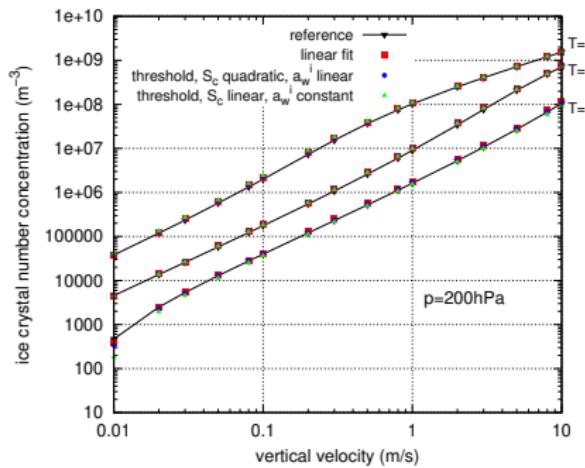


## Change in linear approximation

- ▶ Change of linear slope
- ▶ Change of absolute value



## Approximations

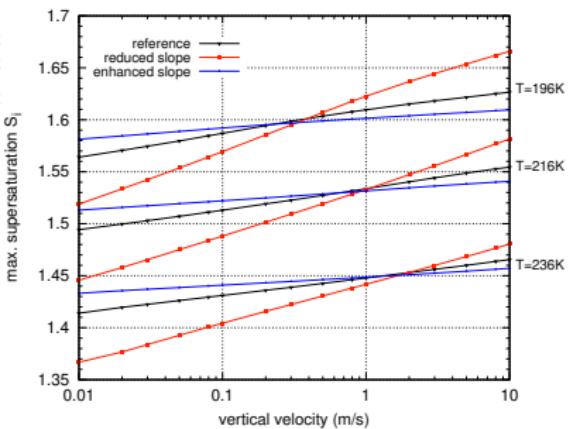
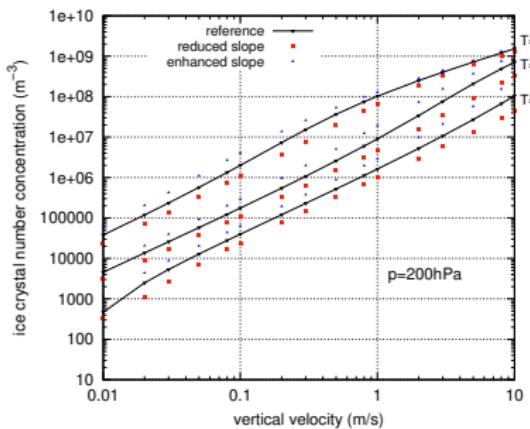
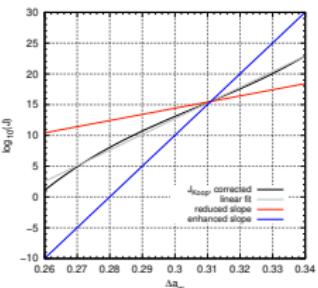


Almost no change visible, less than 15% deviation in number concentrations

→ use  $J = J_0 \exp(a_0 + a_1 \Delta a_w)$  for analysis

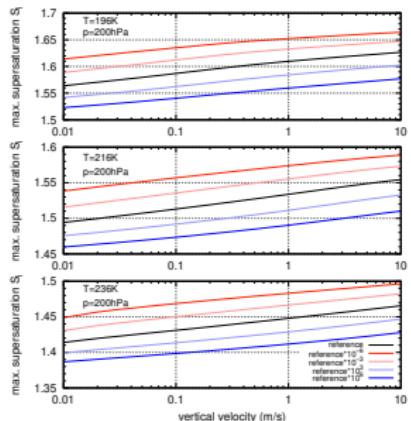
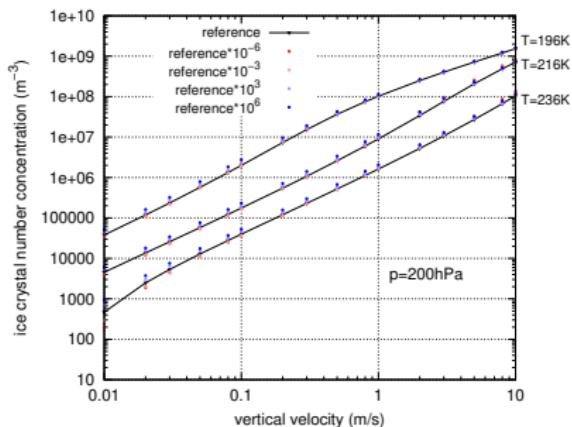
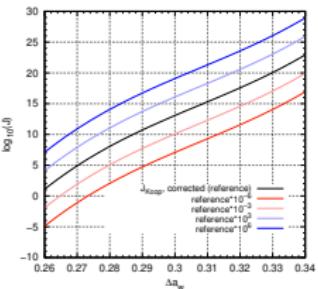
# Change of slope

$$J(\Delta a_w) = J_0 \exp(a_0 + a_1 \Delta a_w) \quad (6)$$

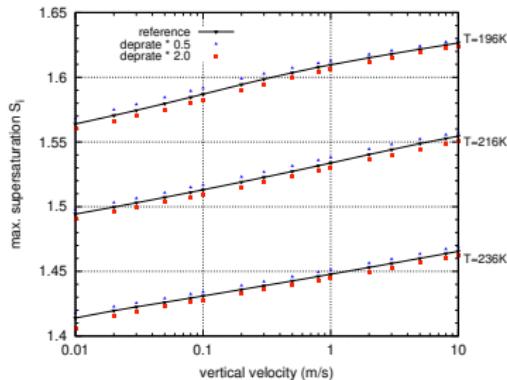
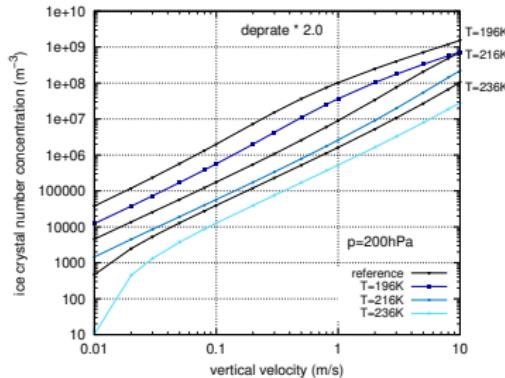
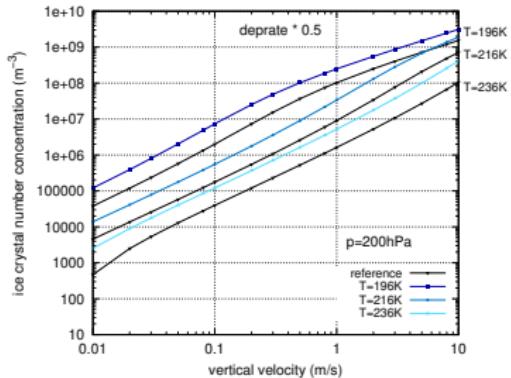


# Change of absolute value

$$J(\Delta a_w) = J_0 \exp(a_0 + a_1 \Delta a_w) \quad (7)$$



# Change in diffusional growth (factor $0.5 \leq f \leq 2$ )



## Investigation of impact of nucleation rate and diffusional growth on idealised nucleation events

- ▶ Nucleation rate can be approximated by linear function
- ▶ Slope of nucleation rate is important
- ▶ Absolute value of nucleation rate only affects maximum supersaturation but only marginally ice crystal number concentrations
- ▶ Diffusional growth has crucial impact on produced ice crystal number concentrations

**Thank you for your attention**

-  Hoose, C. and O. Möhler (2012). "Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments". In: *Atmospheric Chemistry and Physics* 12, pp. 9817–9854.
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-  Krämer, Martina et al. (2016). "A microphysics guide to cirrus clouds - Part 1: Cirrus types". In: *Atmospheric Chemistry and Physics* 16.5, pp. 3463–3483.
-  Luebke, Anna E. et al. (2016). "The origin of midlatitude ice clouds and the resulting influence on their microphysical properties". In: *Atmospheric Chemistry and Physics* 16.9, pp. 5793–5809.
-  Spichtinger, P. and K. M. Gierens (2009). "Modelling of cirrus clouds – Part 1a: Model description and validation". In: *Atmospheric Chemistry and Physics* 9.2, pp. 685–706.