A sampling of recent progress toward understanding ice nucleation and its effects on clouds

R. A. Shaw

Atmospheric Sciences Program & Dept. of Physics

Michigan Technological University

Houghton, MI

A review of work from many groups;

Acknowledgement for my work: US DOE Atmospheric System Research Program

The challenge of ice nucleation parameterization…

Atmos. Chem. Phys., 12, 12061–12079, 2012
www.atmos-chem-phys.net/12/12061/2012/
doi:10.5194/acp-12-12061-2012

Atmospheric Chemistry and Physics

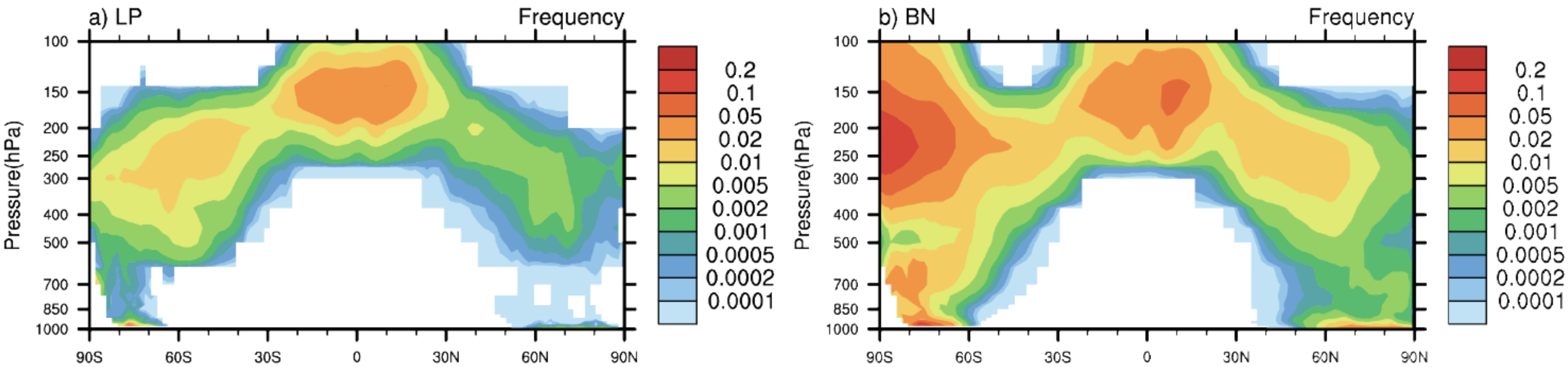
Sensitivity studies of dust ice nuclei effect on cirrus clouds with the Community Atmosphere Model CAM5

X. Liu[1], X. Shi[1,2], K. Zhang[1], E. J. Jensen[3], A. Gettelman[4], D. Barahona[5,6], A. Nenes[7,8], and P. Lawson[9]

[1]Atmospheric Science & Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA
[2]Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China
[3]NASA Ames Research Center, Moffett Field, CA, USA
[4]National Center for Atmospheric Research, Boulder, CO, USA
[5]Global Modeling and Assimilation Office, NASA Goddard Space Flight Center, Greenbelt, MD, USA
[6]I. M. Systems Group, Rockville, MD, USA
[7]School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA, USA
[8]School of Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, GA, USA
[9]SPEC Inc., Boulder, CO, USA

- Investigation of influence of two different parameterizations of homogeneous and heterogeneous ice nucleation.
- The parameterizations differ significantly in the number concentration of dust ice nuclei.

The challenge of ice nucleation parameterization…



- Frequency distribution of homogeneous nucleation events in CAM5 for two different ice nucleation parameterizations.
- The differences arise from the competition between heterogeneous and homogeneous ice nucleation.

Liu et al., ACP 2012

Immersion freezing described by water activity...

PAPER 165/24 www.rsc.org/faraday_d | Faraday Discussions

A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets†

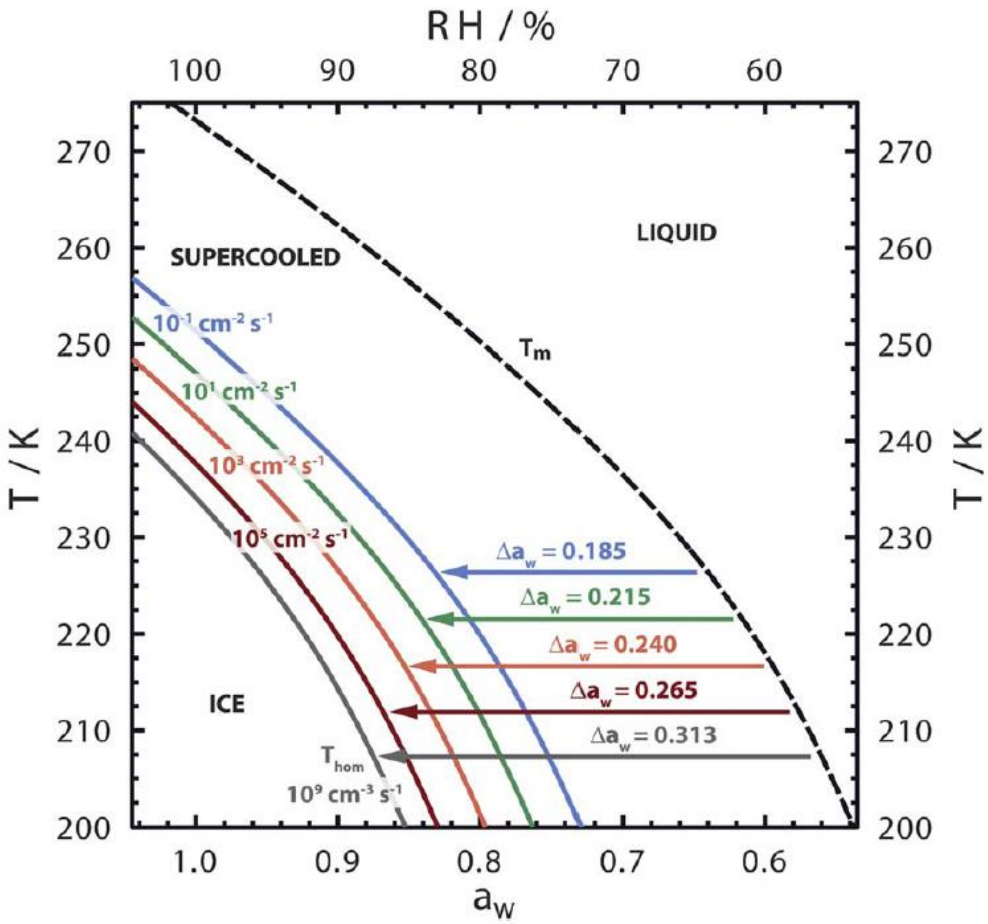
Daniel A. Knopf* and Peter A. Alpert

Received 8th March 2013, Accepted 23rd April 2013
DOI: 10.1039/c3fd00035d

- Activity-based immersion freezing model unites effects of solute concentration, ice nucleus surface area, and cooling rate (time).
- Provides the basis for a robust, physically-based parameterization of heterogeneous ice nucleation.
- Homogeneous and heterogeneous ice nucleation are not tied together as, for example, with parameterization of Kaercher & Lohmann.

Question: Is this level of detail realistically able to be included in cloud models, especially for heterogeneous aerosol populations?

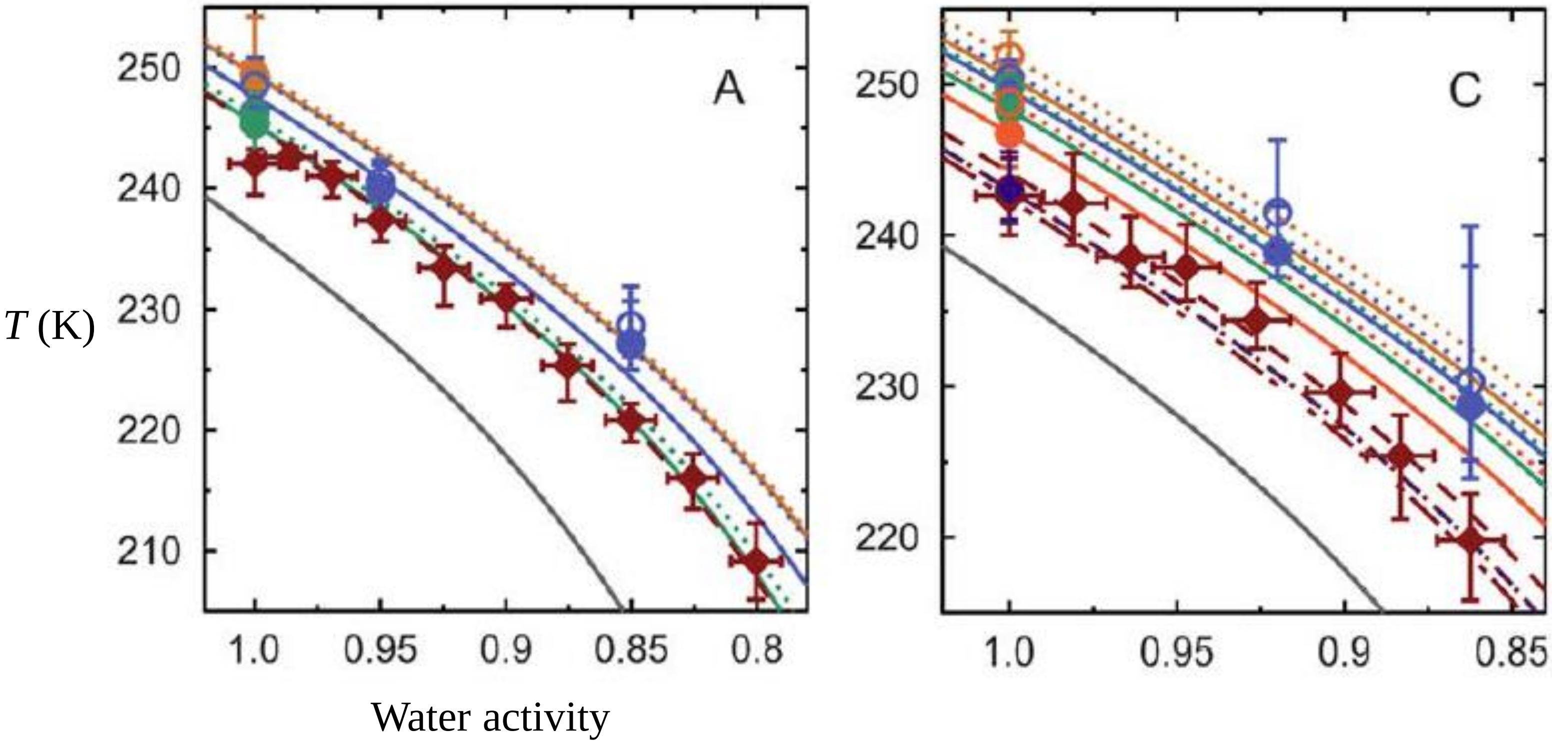
Immersion freezing described by water activity...



Based on classical nucleation theory; extension of activity parameterization for homogeneous nucleation.

Knopf & Alpert, Faraday Discuss. 2013

Immersion freezing described by water activity...



The approach is validated through extensive measurements with mineral dusts, biological particles, and organic compounds.

Knopf & Alpert, Faraday Discuss. 2013

"Mixed" stochastic models of ice nucleation…

E.g., soccer ball model

It breaks all implicit connections between the number of patches, the size of the patches, and the distribution of nucleation rates.

It is therefore general, and allows for various idealized limits:

- $\sigma_\theta = 0 \rightarrow$ Uniform population, i.e., the traditional, purely stochastic view.
- $n = 1$ and $\sigma_\theta > 0 \rightarrow$ Externally mixed population.
- $n \gg 1$ and $\sigma_\theta > 0 \rightarrow$ Internally mixed population (with statistically identical particles).

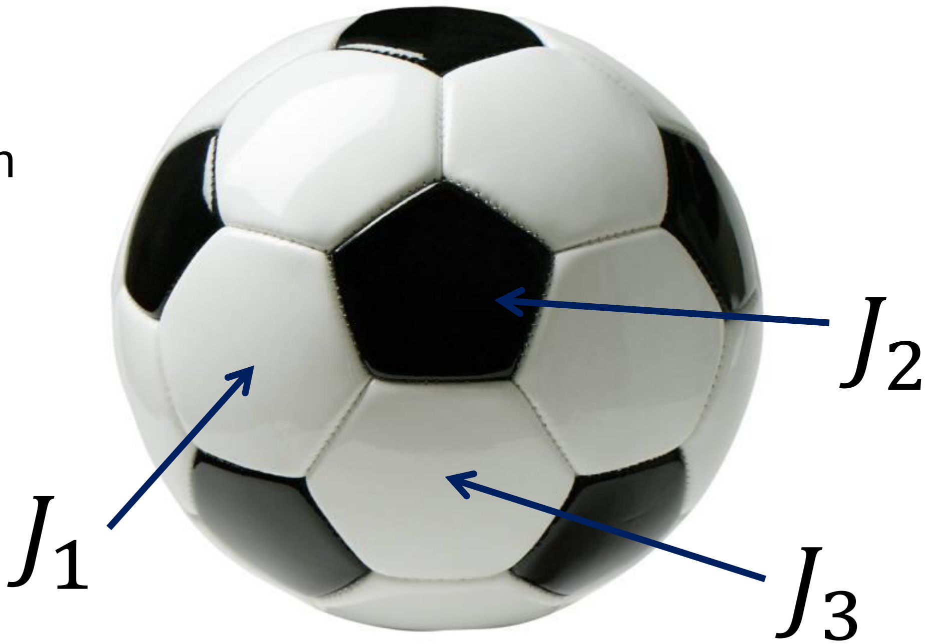
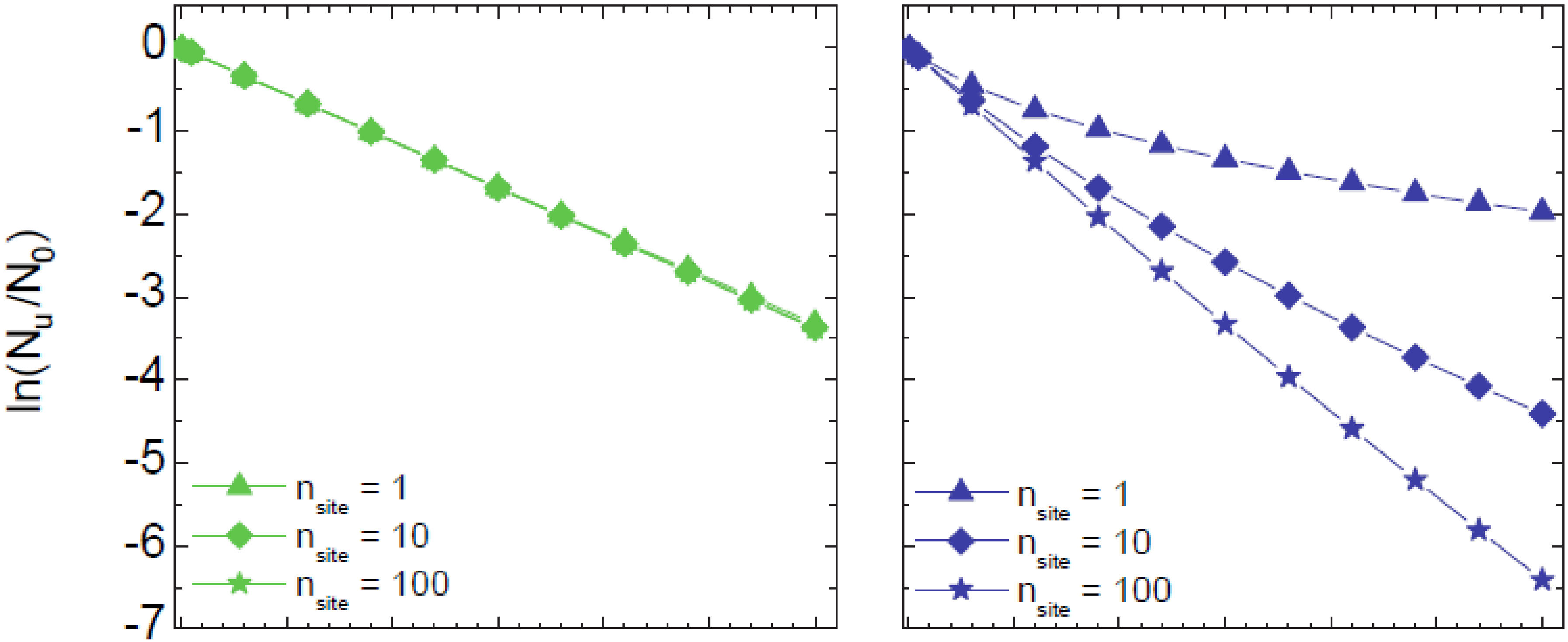


Image from regentsprep.org

Niedermeier et al., ACP 2010

Emergence of "singular" behavior from a purely stochastic model



$\sigma_\theta = 0.001$ rad

$\sigma_\theta = 0.01$ rad

Probability of "survival" (not freezing) versus time:

- Narrow θ-distribution $\rightarrow$ exponential decay, purely stochastic.
- Broad θ-distribution, small n $\rightarrow$ decay tends to saturate, seemingly singular.

Niedermeier et al., ACP 2010

Ice nucleation on biological particles...

Suspendable macromolecules are responsible for ice nucleation activity of birch and conifer pollen

B. G. Pummer[1], H. Bauer[2], J. Bernardi[3], S. Bleicher[4], and H. Grothe[1]

[1] Institute of Material Chemistry, Vienna University of Technology, Austria
[2] Institute of Chemical Technologies and Analytics, Vienna University of Technology, Austria
[3] USTEM, Vienna University of Technology, Austria
[4] Atmospheric Chemistry Research Laboratory, University of Bayreuth, Germany

Immersion freezing of birch pollen washing water

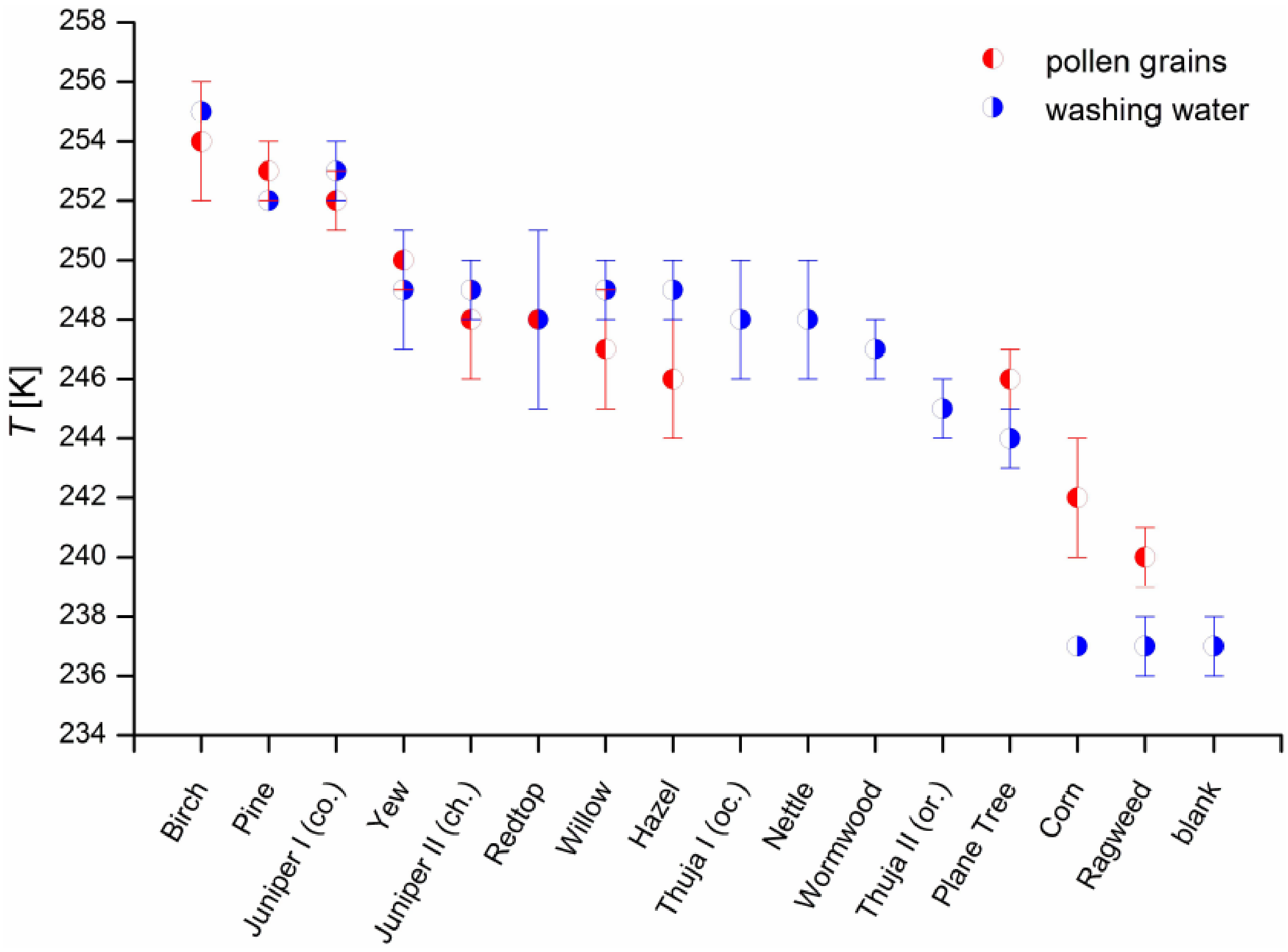
S. Augustin[1], H. Wex[1], D. Niedermeier[1], B. Pummer[2], H. Grothe[2], S. Hartmann[1], L. Tomsche[1], T. Clauss[1], J. Voigtländer[1], K. Ignatius[1], and F. Stratmann[1]

[1] Leibniz Institute of Tropospheric Research, Permoserstr. 15, 04318 Leipzig, Germany
[2] Institute of Material Chemistry, Vienna University of Technology, Austria

Ice nucleation efficiency for some pollens comes from macromolecules that can be suspended in washing water.
Question: Could this be a route for multiplying the biological effect beyond pollen particle concentrations?

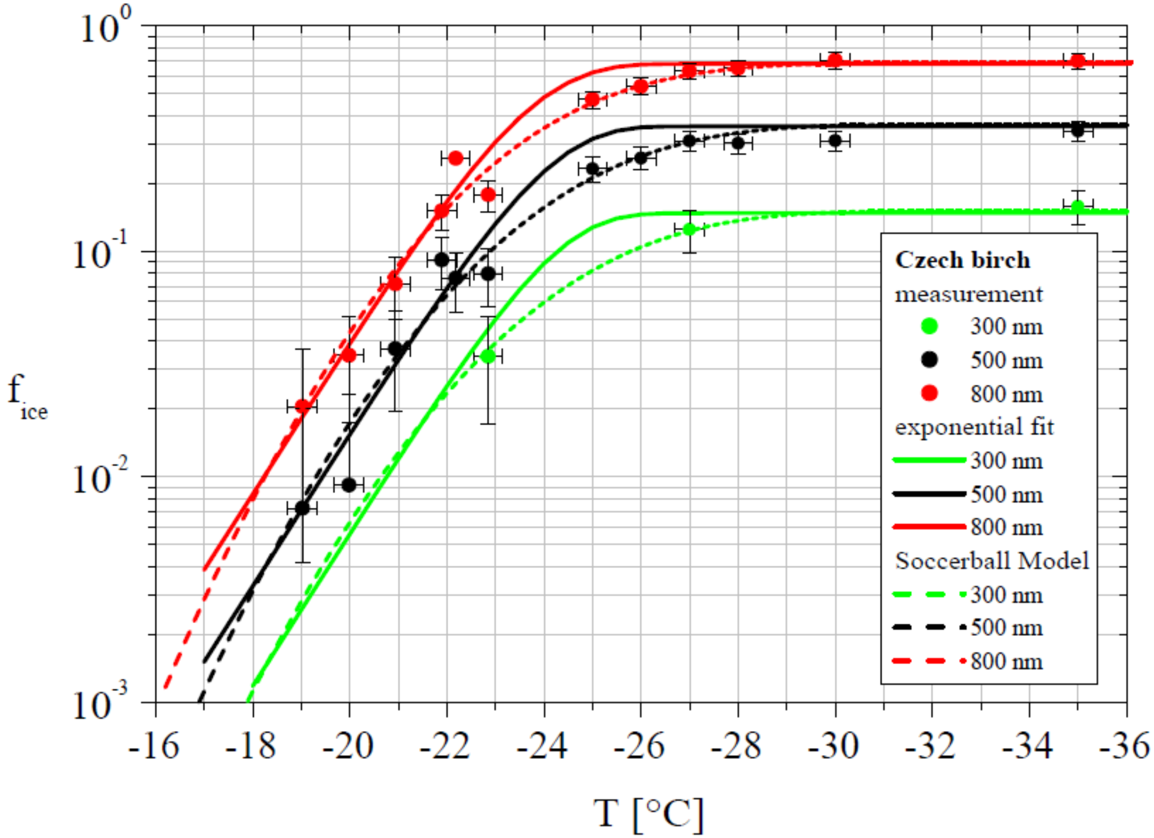
Pummer et al., Atmos. Chem. Phys. 2012
Augustin et al., Atmos. Chem. Phys. accepted

Biological ice nuclei...



Pummer et al., Atmos. Chem. Phys. 2012

Biological ice nuclei...



- Ice nucleation by macromolecules obtained from birch pollen.
- The observed plateaus in freezing probability allow ice nucleation rate for single macromolecules to be measured.

Augustin et al., Atmos. Chem. Phys. 2013

Depletion of ice nuclei...

A FIRE-ACE/SHEBA Case Study of Mixed-Phase Arctic Boundary Layer Clouds: Entrainment Rate Limitations on Rapid Primary Ice Nucleation Processes

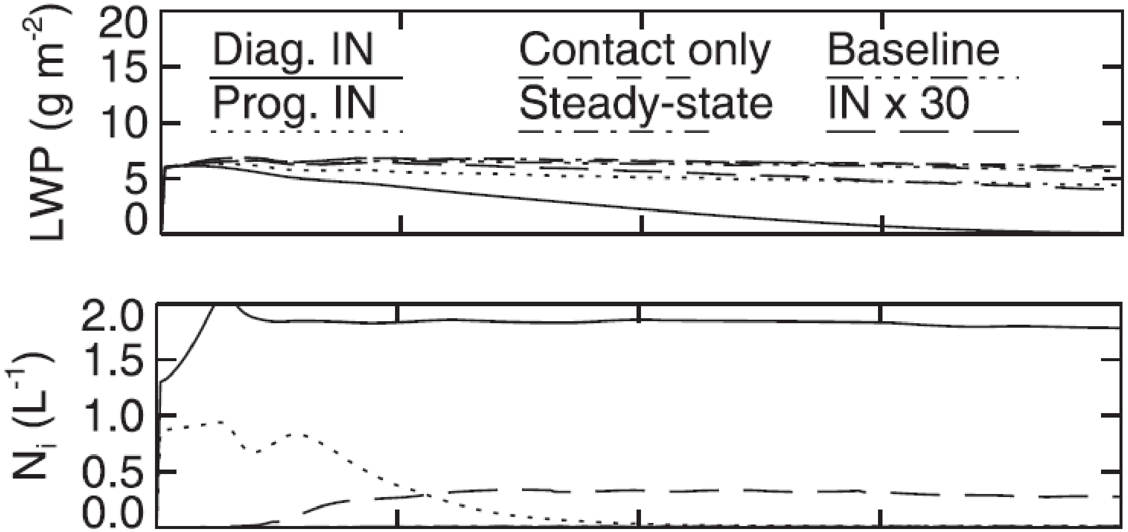
ANN M. FRIDLIND,[*] BASTIAAN VAN DIEDENHOVEN,[+] ANDREW S. ACKERMAN,[*]
ALEXANDER AVRAMOV,[+] AGNIESZKA MROWIEC,[+] HUGH MORRISON,[#]
PAQUITA ZUIDEMA,[@] AND MATTHEW D. SHUPE[&]

[*] *NASA Goddard Institute for Space Studies, New York, New York*
[+] *Columbia University, New York, New York*
[#] *National Center for Atmospheric Research, Boulder, Colorado*
[@] *Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, Florida*
[&] *Cooperative Institute for Research in Environmental Sciences, University of Colorado, and NOAA/ESRL/PSD, Boulder, Colorado*

Arctic stratus & altocumulus clouds tend to be thin, long-lived, with weakly precipitating ice.

Question: Where do all the ice nuclei come from?!

Depletion of ice nuclei...



$$H\frac{dN_i}{dt} = w_e N_{\mathrm{IN}} - (\upsilon_f + w_e)N_i. \qquad (1)$$

For the simulated conditions, $w_e \ll \upsilon_f$ (see Table 3), and Eq. (1) can be simplified to

$$H\frac{dN_i}{dt} = w_e N_{\mathrm{IN}} - \upsilon_f N_i. \qquad (2)$$

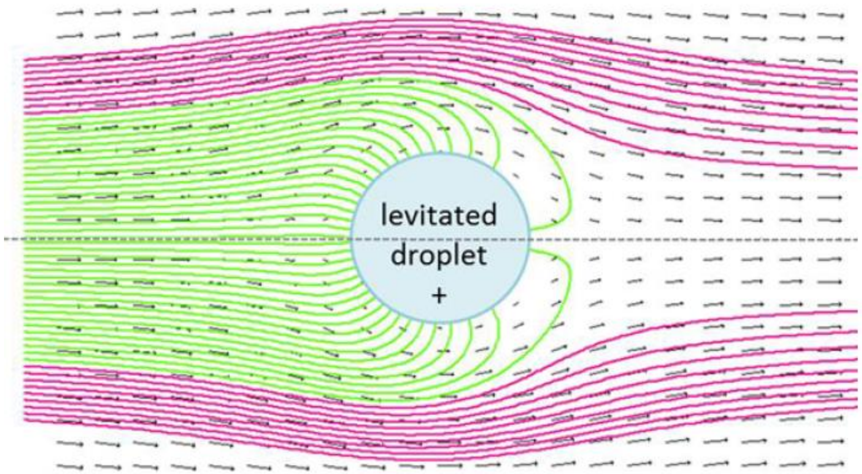
Dividing the ice crystal reservoir HN_i by its sink $\upsilon_f N_i$ gives an e-folding time scale H/υ_f of about 20–30 min on which N_i relaxes toward its steady-state value

$$N_i = N_{\mathrm{IN}} w_e / \upsilon_f. \qquad (3)$$

- Entrainment speed is much less than ice crystal fall speed.
- Ice concentration is much less than the ice nucleus concentration.

Fridlind et al., JAS 2012

Contact nucleation...



Atmos. Meas. Tech., 6, 2373–2382, 2013
www.atmos-meas-tech.net/6/2373/2013/
doi:10.5194/amt-6-2373-2013

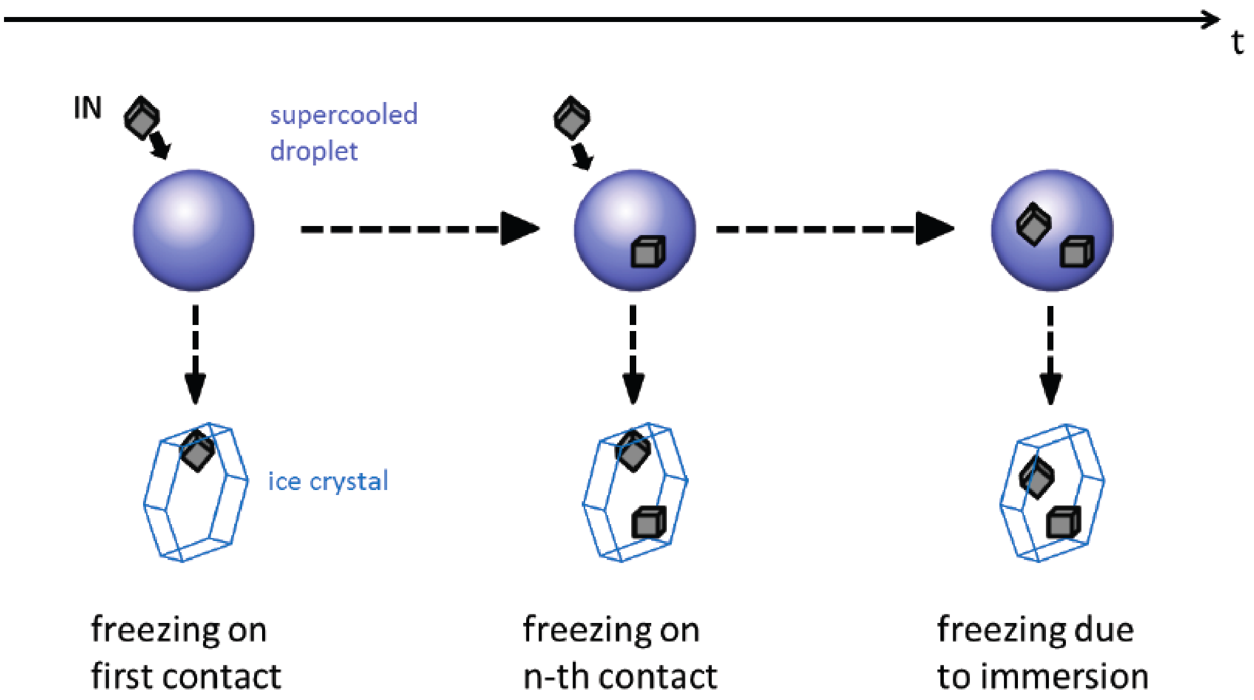
Experimental quantification of contact freezing in an electrodynamic balance

N. Hoffmann, A. Kiselev, D. Rzesanke, D. Duft, and T. Leisner
Karlsruhe Institute of Technology, Mailbox 3640, 76021 Karlsruhe, Germany

- A supercooled water droplet is suspended in an electrodynamic balance.
- A steady air flow containing insoluble particles results in collisions between the particles and the droplet.

See also Hoffmann et al., Faraday Discuss. 2013

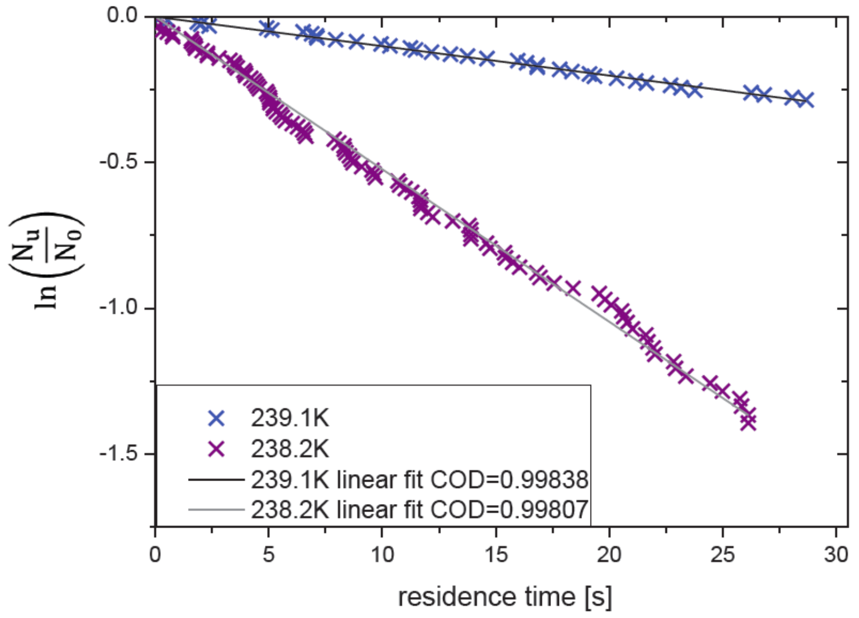
Contact nucleation...

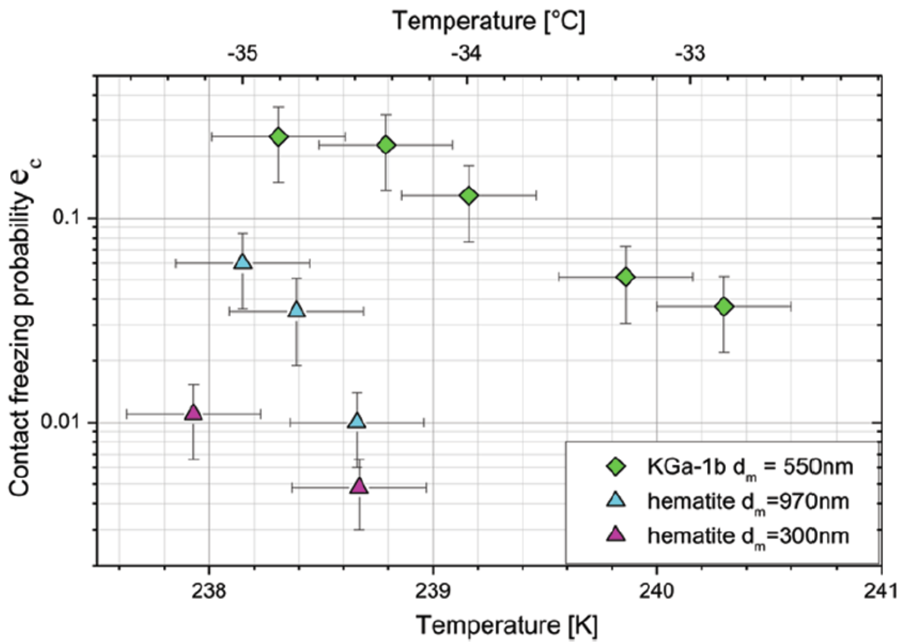


- The droplet can freeze upon contact, or upon immersion after contact.

Hoffmann et al., Atmos. Meas. Tech. 2013

Contact nucleation...

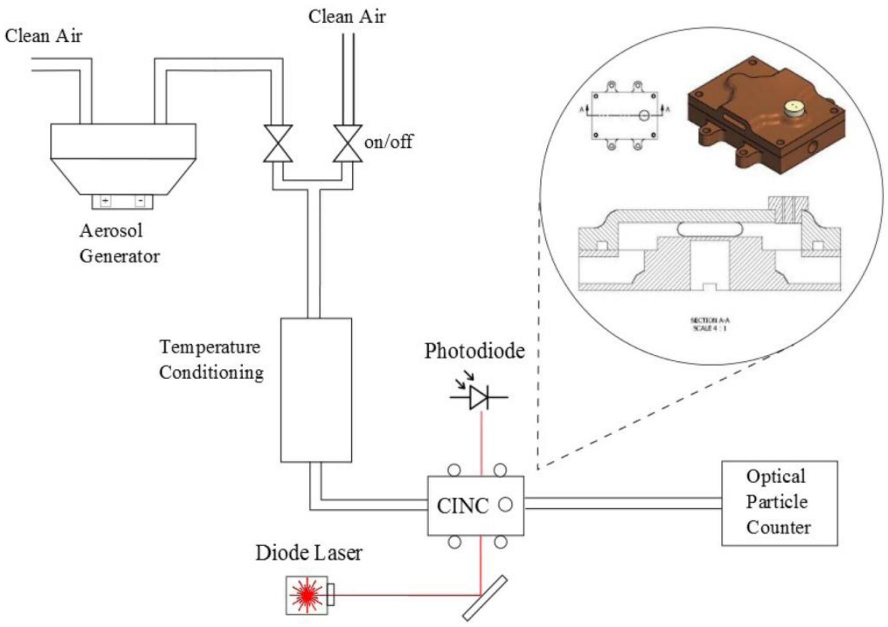


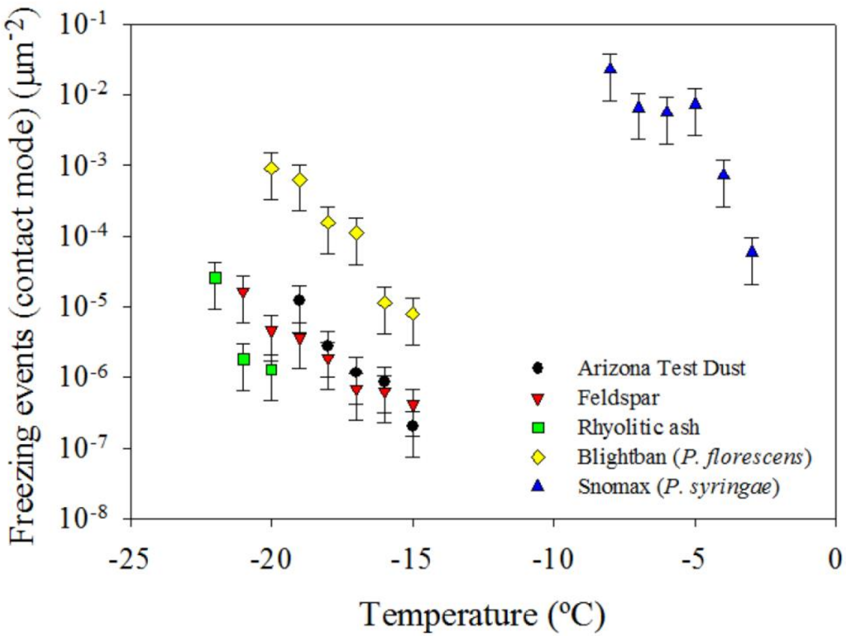


- Freezing rate is equal to product of the collision rate and the probability of freezing on a single contact.
- Collision rate can be calculated (and confirmed by independent measurements, so measured exponential survival curve results in the single-contact freezing probability.

Hoffmann et al., Atmos. Meas. Tech. 2013

Contact nucleation...





- Methods are finally being developed for quantifying contact nucleation.
- The freezing efficiency in this experiment is defined as the ratio of the number of freezing events to the total aerosol surface area coming in contact with the supercooled drop.

Niehaus, Cantrell, et al., JAOTech in review

Some unanswered questions for future research…

- How relevant is the “stochastic” aspect of ice nucleation in the atmosphere? Are singular approximations good enough?
- Can parameterizations based on classical nucleation theory (e.g., activity-based) be combined with internally-mixed models (e.g., soccer ball) to yield useful parameterizations?
- What is the role of biological ice nuclei? Can we achieve an understanding of the “active” macromolecules?
- What is the explanation for long-lived ice precipitation from stratiform, mixed-phase clouds? What is the source of ice nuclei?
- What is the physical basis for contact nucleation? Can it be quantified and included in models?