What processes control ice nucleation and its impact on ice-containing clouds ?

Xiaohong Liu (PNNL)

With thanks to: P. DeMott (CSU),
G. Kulkarni, S. Burrows (PNNL)

Pacific Northwest NATIONAL LABORATORY

Challenges

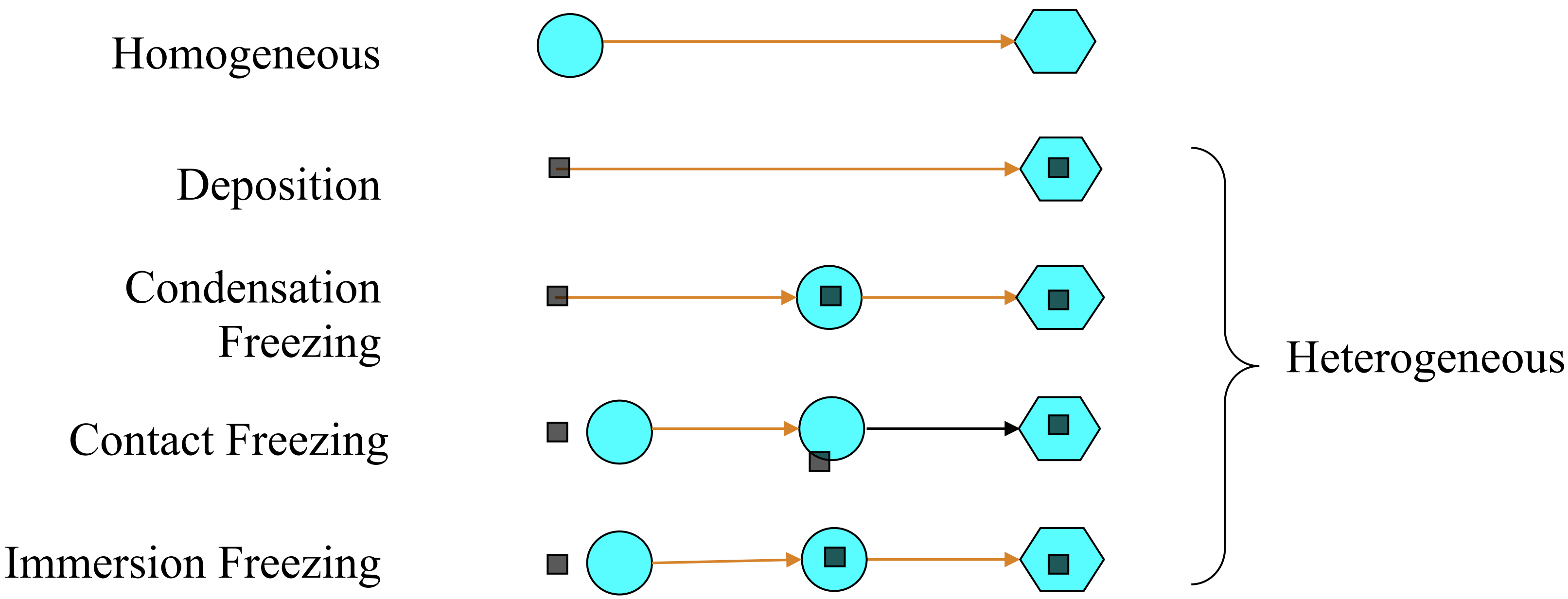
- Ice nucleation processes involving aerosols are key to the formation and properties of cirrus and mixed-phase clouds, and thereby can impact both the atmospheric radiative energy distribution and precipitation processes.

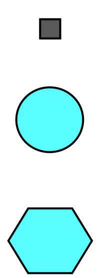
- Compared to droplet formation in warm clouds, ice nucleation is more complicated and much less understood.

- Large uncertainties exist in the representation of ice nucleation processes in climate models, and aerosol effects on mixed-phase and cirrus clouds.

How ice crystals are formed?

<u>Multiple Ice Nucleation Mechanisms</u>





Soluble/insoluble aerosol particle (substrate) (~10^{-3} – 10^{-5} of aerosol population)

Supercooled solution droplet / cloud droplet

Ice crystal

Courtesy of G. Kulkarni

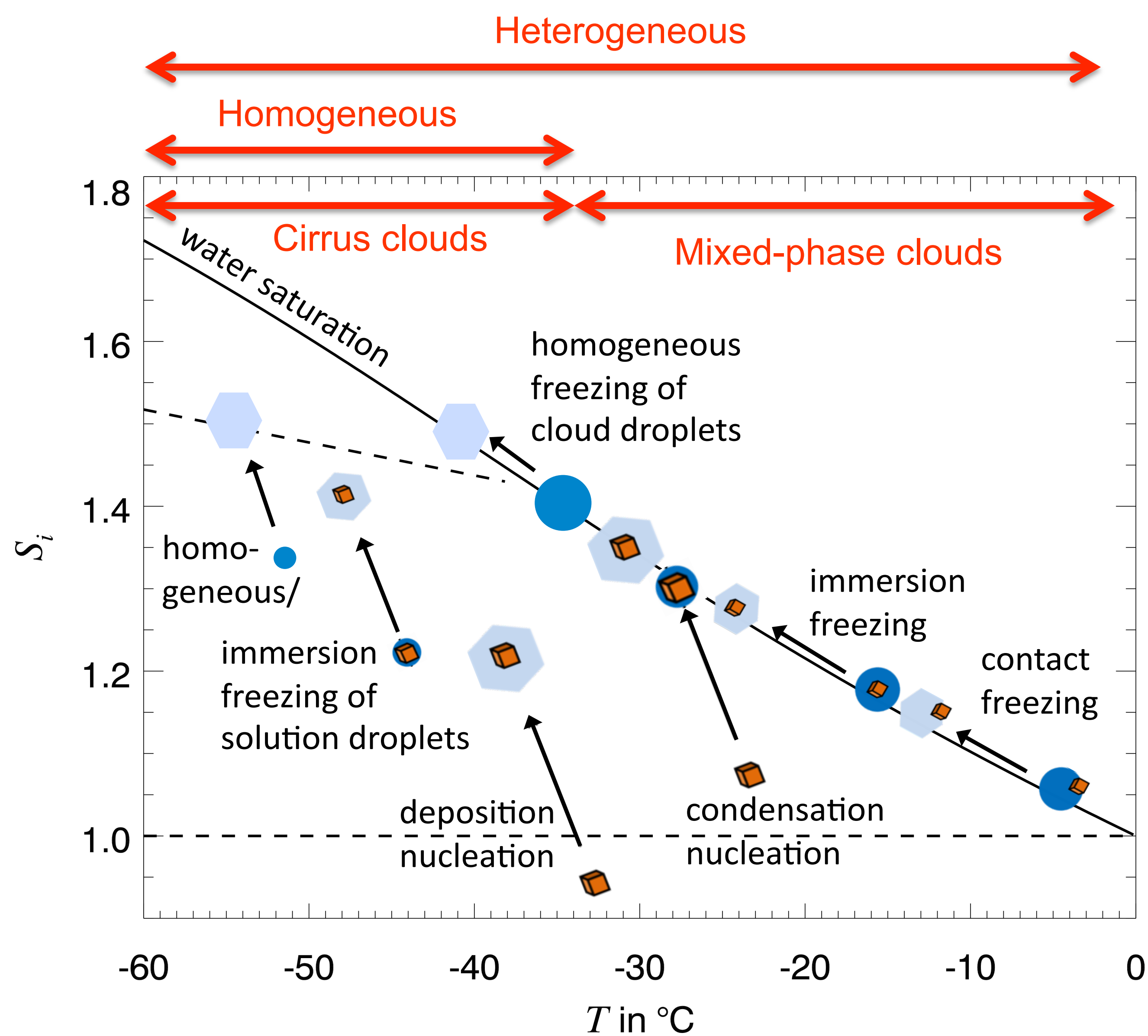
Pacific Northwest NATIONAL LABORATORY

Modes of ice nucleation

No clouds
without aerosol
particles
➔ CCN

No clouds
without aerosol
particles
➔IN (ice nuclei)

Except freezing
of water droplets
at -36 °C



Hoose and Moeller, ACPD (2012)

Science Questions

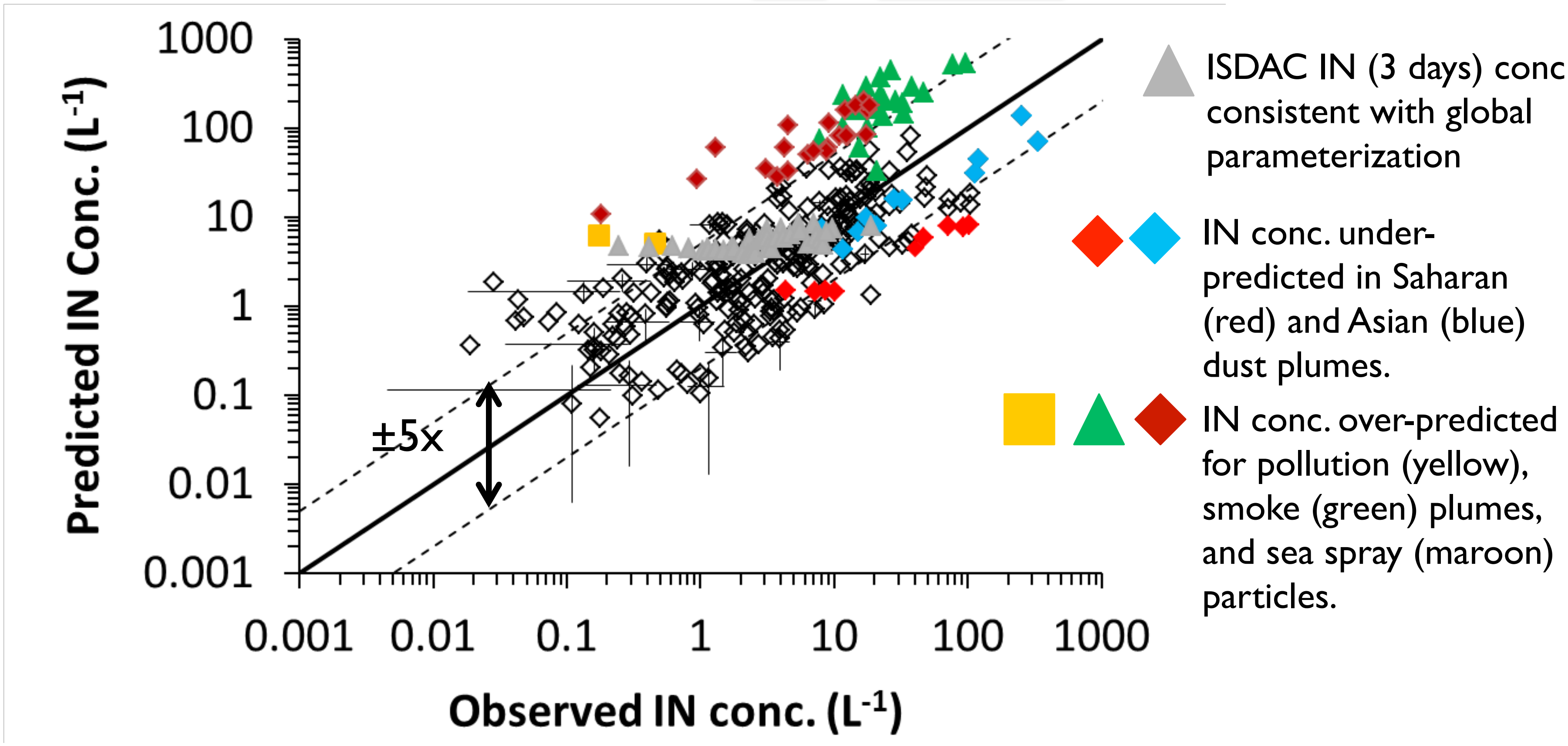
1. How does ice nucleation depend on aerosol properties, environmental conditions, and time?
2. What are the specific contributions of mineral dust, soot, organics, and biological aerosols toward ice nucleation and IN variability?
3. What measurement approaches are needed to characterize all relevant ice nucleation processes?
4. Can we reach IN-ice crystal closure at current stages of IN and ice crystal measurement? If not, what areas are needed for improvement?
5. What are the roles of ice nucleation on cloud and precipitation properties and climate forcing?

Q1. How does ice nucleation depend on aerosol properties, environmental conditions, and time?

- Aerosol properties
  - Size, morphology, composition, surface coating
- Environmental conditions
  - Temperature,
  - Relative humidity
  - Updraft (cooling rate)
- Time dependence
  - Stochastic vs. singular for heterogeneous nucleation

Ice nucleation in mixed-phase clouds

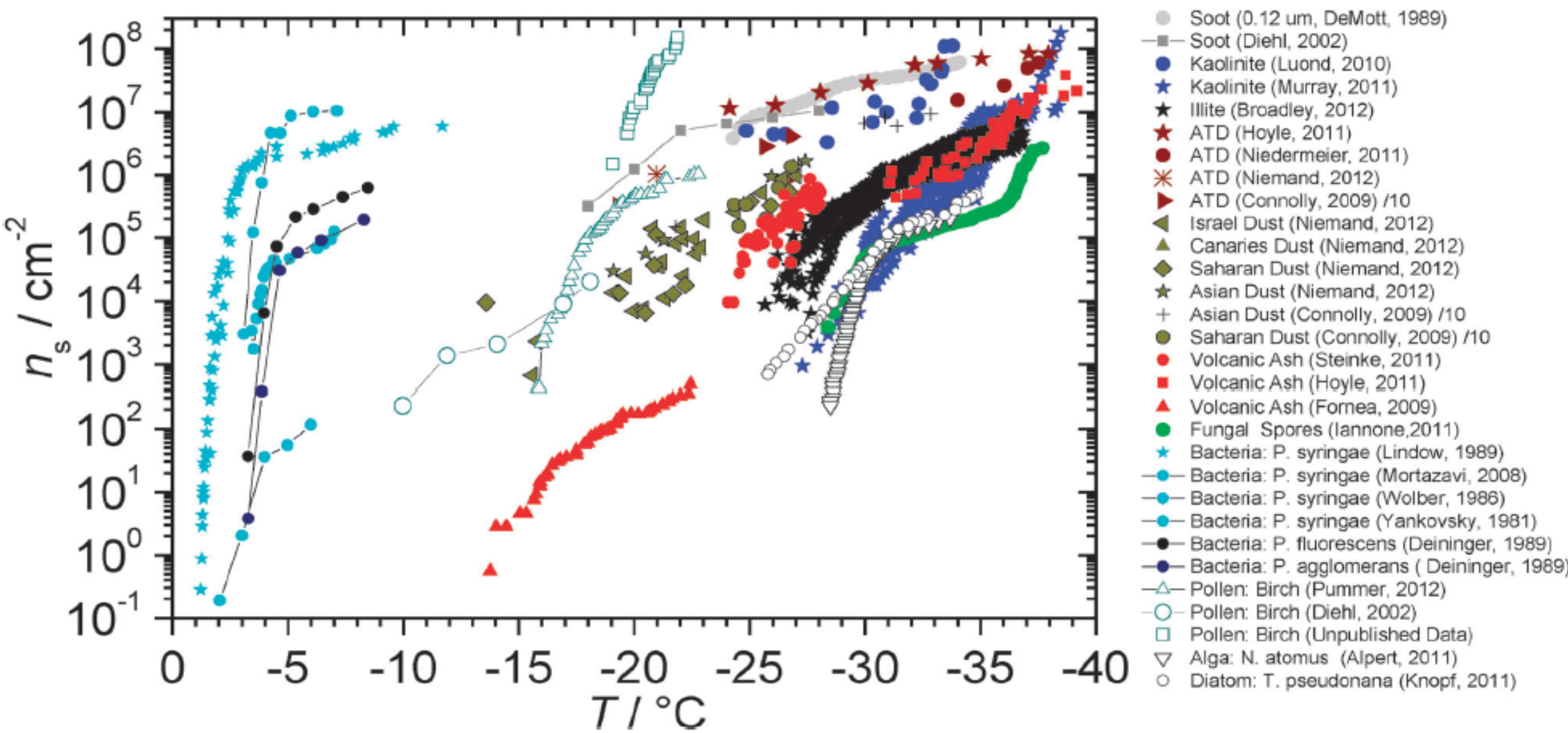
$$n_{IN,T_k} = a(273.16 - T_k)^b (n_{aer,0.5})^{(c(273.16 - T_k)+d)}$$



DeMott et al. (2010, PNAS) parameterization: T and $n_{aer,0.5}$

Composition matters as well

Immersion freezing based on aerosol surface active site density

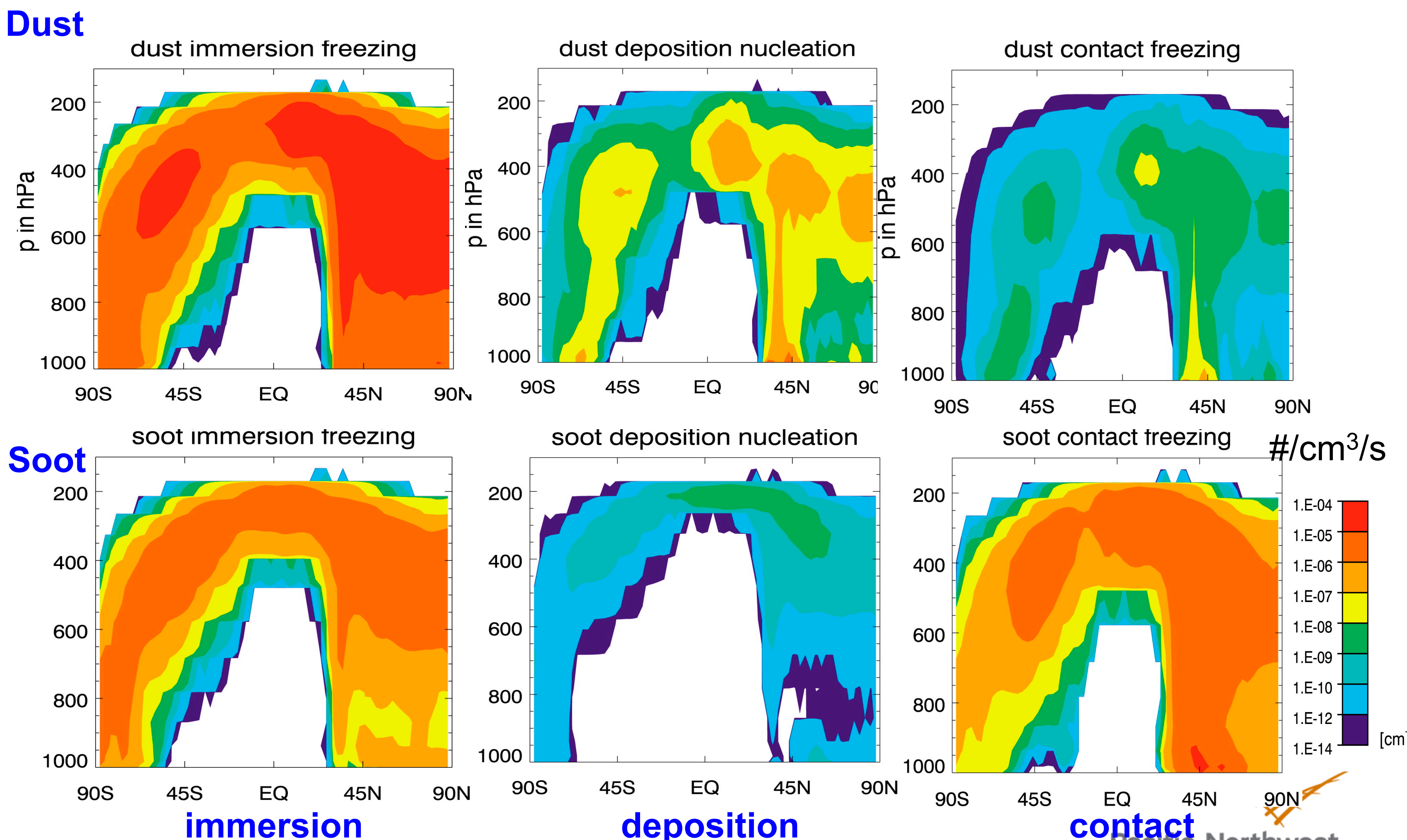


Immersion freezing efficiency depends upon the aerosol surface area and chemistry.
In general, for the same aerosol size ice nucleating properties depend upon the aerosol chemistry.

Pacific Northwest
NATIONAL LABORATORY

Murray et al. 2012

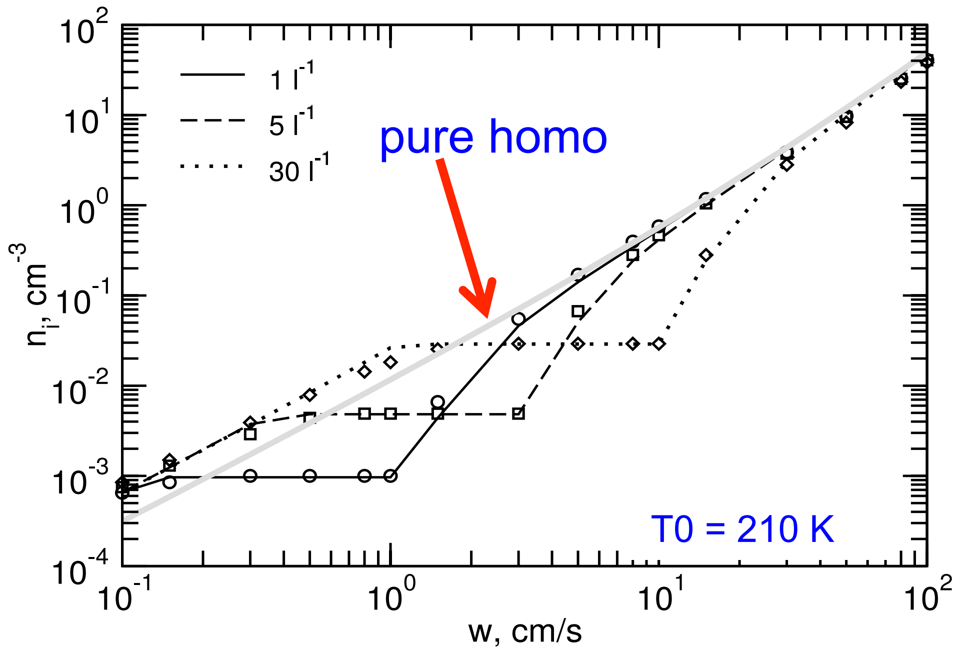
Ice nucleation in mixed-phase clouds: immersion vs. deposition vs. contact mode



Pacific Northwest NATIONAL LABORATORY

CAM-Oslo, Hoose et al, JAS 2010

Ice nucleation in cirrus clouds (T< -37C): competition of homogeneous (on sulfate) and heterogeneous (immersion & deposition) modes



Role of vertical updraft (w):

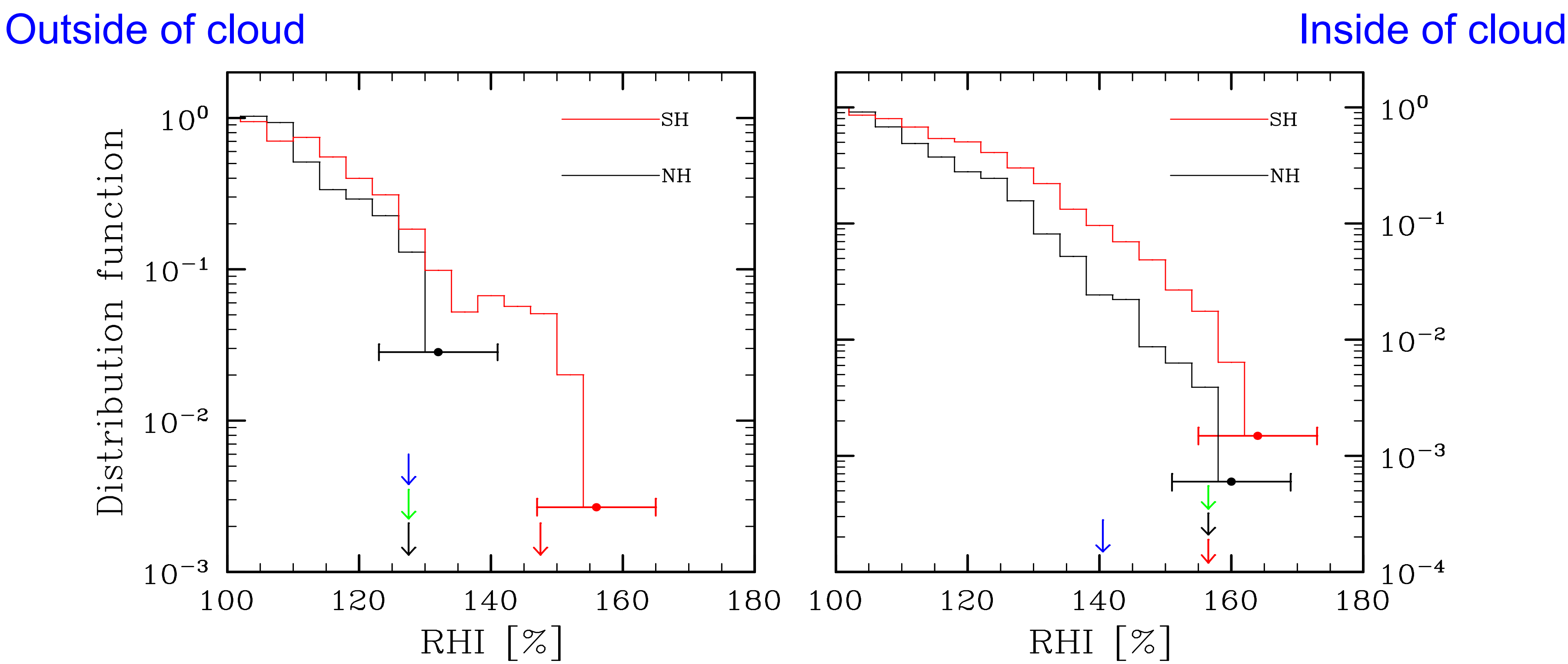
(1) Determining supersaturation in clouds and thus relative importance of homogeneous vs. heterogeneous nucleation

(2) Determining sensitivity of Ni to aerosol number Na for homogeneous nucleation (larger AIE with higher w)

Karcher et al. (2006)

Pacific Northwest NATIONAL LABORATORY

Ice nucleation in cirrus clouds: competition of homogeneous and heterogeneous (immersion & deposition) modes



PDF(RHi) from observations during the Interhemispheric Differences in Cirrus Properties From Anthropogenic Emissions (INCA) in NH and SH

Haag et al. (2003)

Q2. What are the specific contributions of coated/uncoated mineral dust, volcanic ash, soot, organics, and biological aerosols toward ice nucleation and IN variability?

- Characteristics of nucleation efficiency of aerosol types
  - onset T and RH, surface active site density
  - nucleation mode (e.g., immersion, deposition)

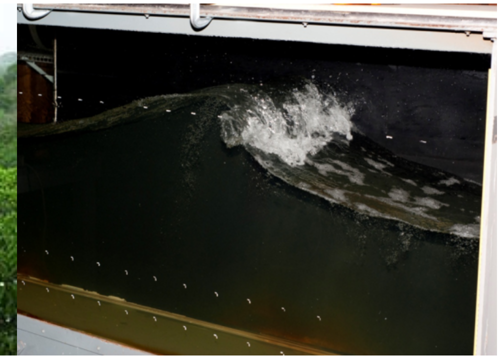
- Effect of surface coating
  - composition dependency
  - coating thickness
  - change of nucleation mode

Types of aerosol as ice nuclei

Smoke: prescribed burns (Longleaf Pine), Newton, GA

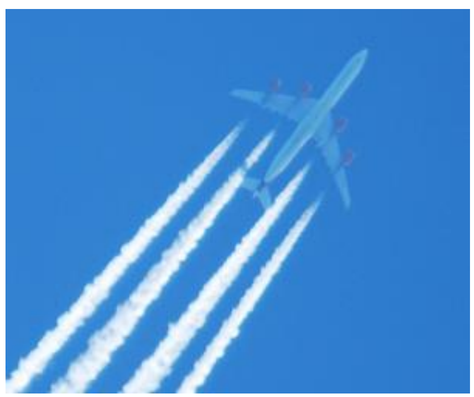
Dust: Saharan Aerosol Layer (SAL)

PBAP (primary biological aerosol particles) dominant in large aerosol, Amazonian Rainforest



Seawater spray: CAICE wave channel, Scripps/ UCSD, CA

Industry Pollution: Lead, others?



Aircraft soot

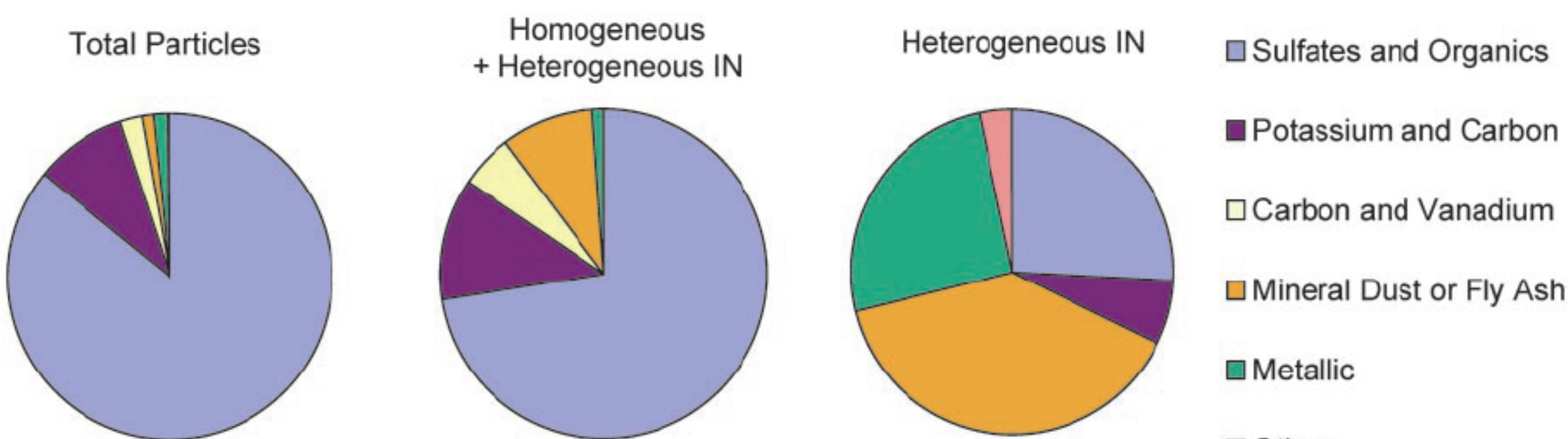
Volcanic ash

Pacific Northwest
NATIONAL LABORATORY

Ice Nuclei

- How do we know what an ice nucleus is?
- From laboratory studies of nucleation rates/thresholds
- From atmospheric measurements of 'residuals' after crystals are evaporated

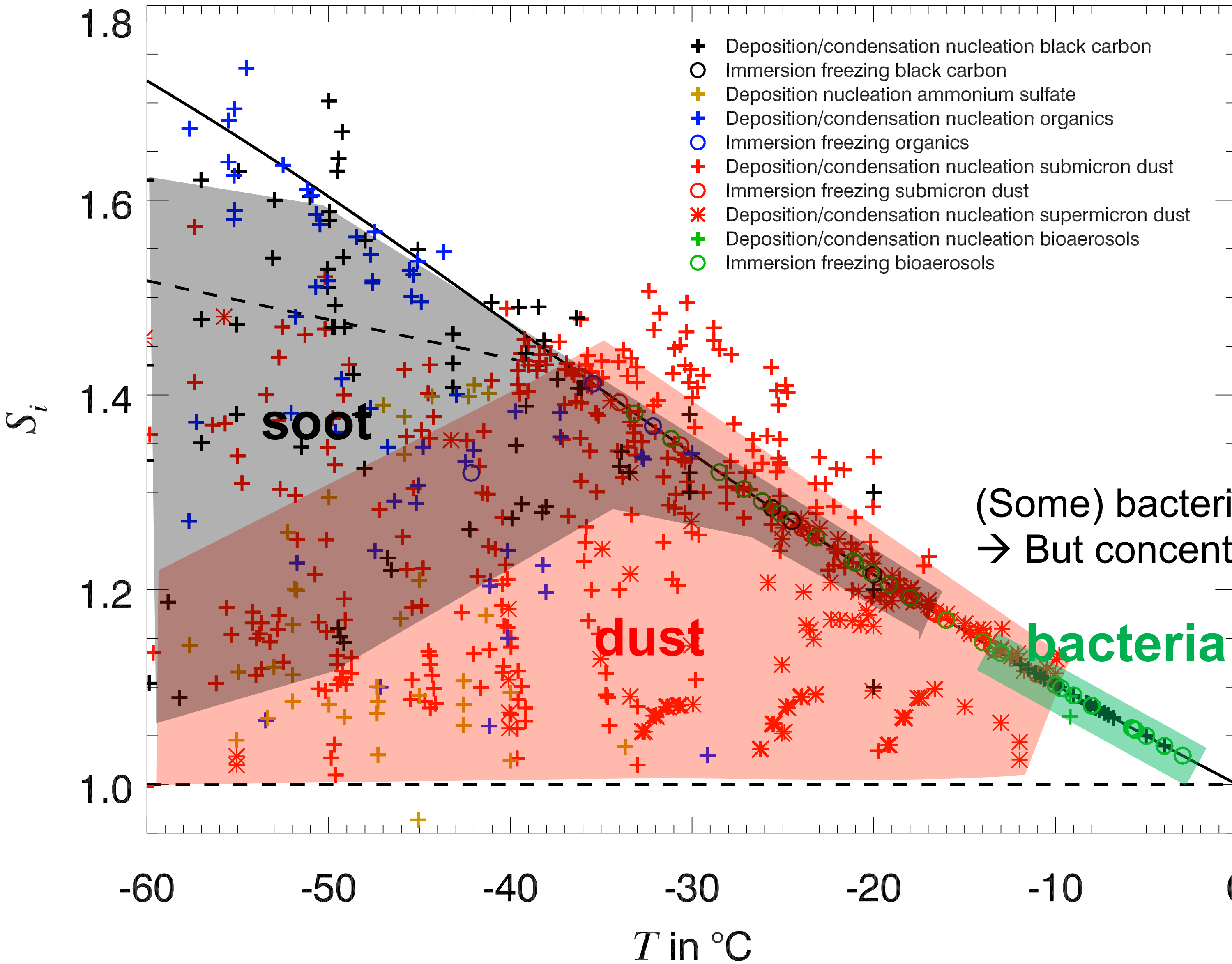
DeMott et al 2003, PNAS



Higher S_{ice} Low S_{ice}

Pacific Northwest NATIONAL LABORATORY

Onset for heterogeneous ice nucleation

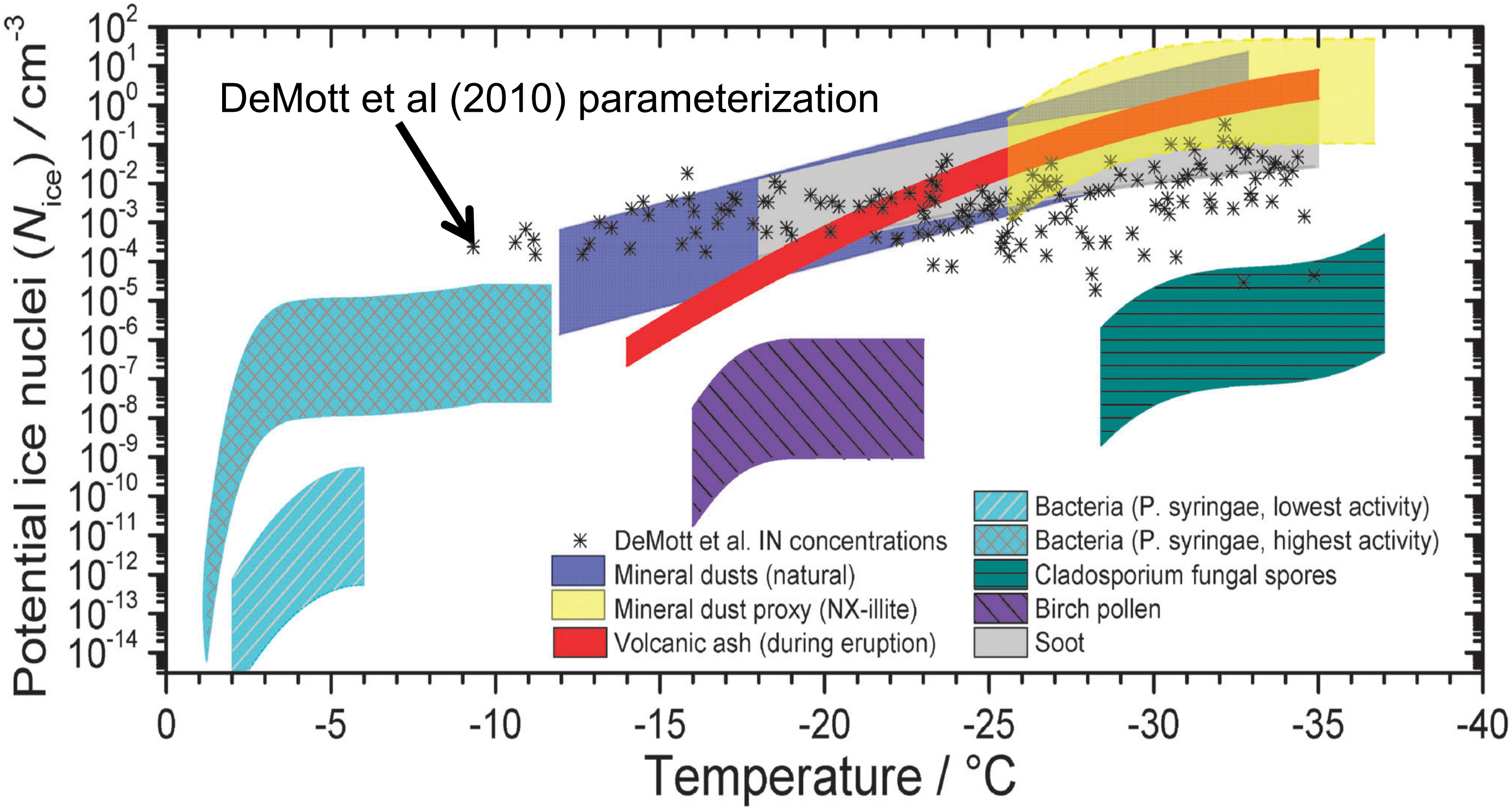


(Some) bacteria are "special"
→ But concentrations are low

Pacific Northwest NATIONAL LABORATORY

Hoose and Möhler, ACP 2012

Immersion mode IN contribution

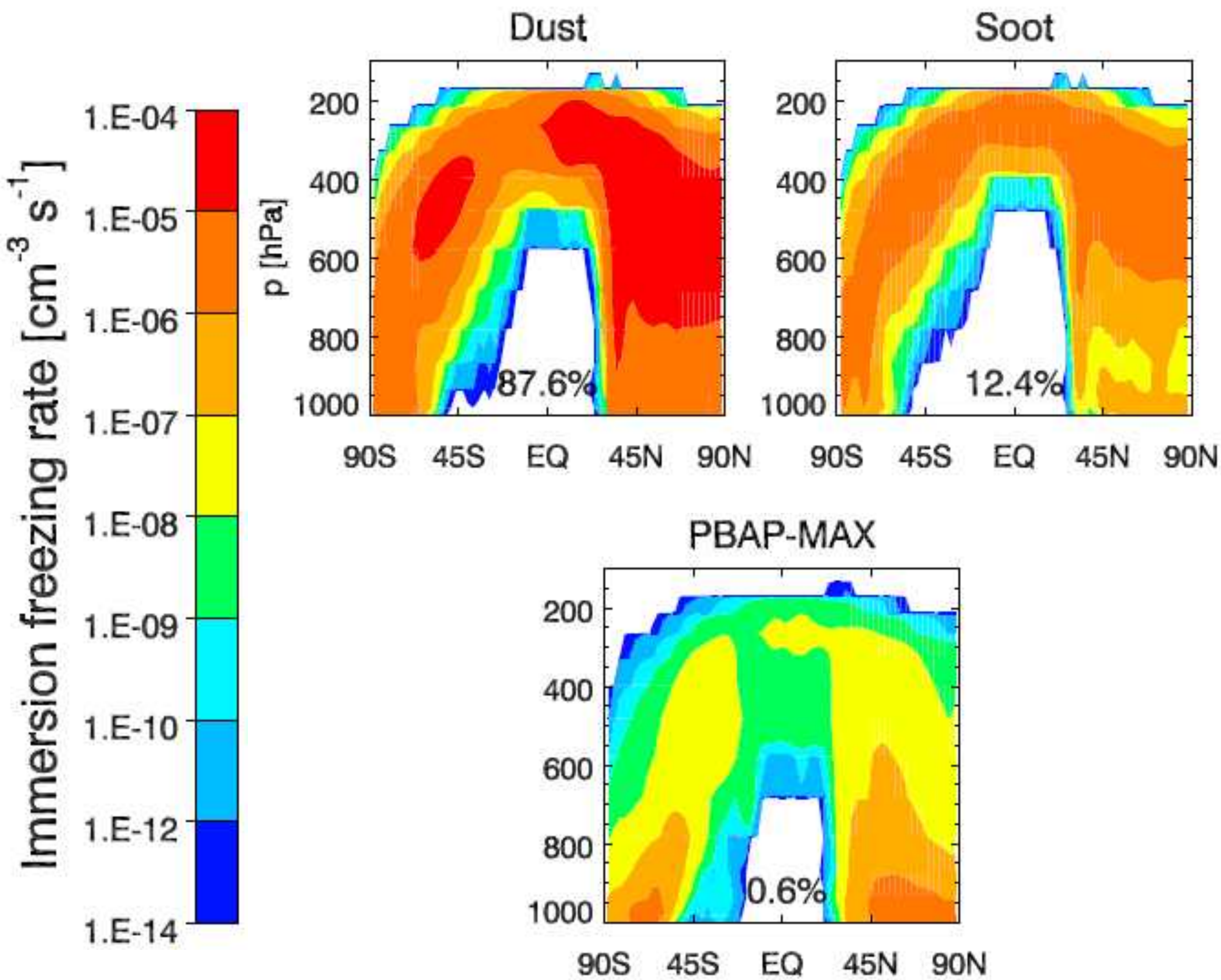


Considerable scatter can be observed among the freezing efficiency of various aerosols.

Mineral dust dominates the potential IN fraction, while bacterial IN concentration is low but effective at warmer temperatures.

Murray et al. 2012

Simulated freezing rate



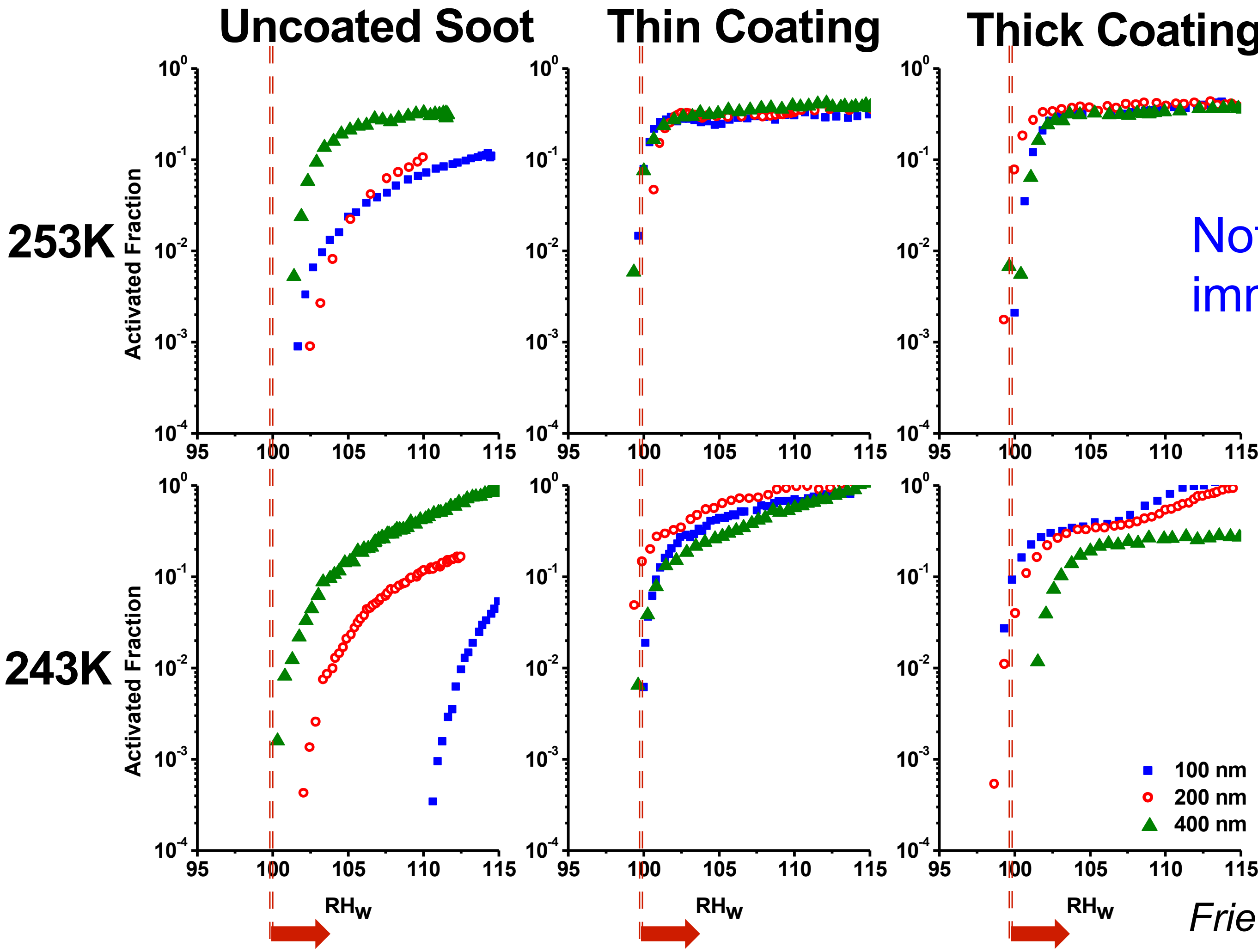
Uppermost estimate:
Mean contribution of 0.6% to global ice nucleation

Generous assumptions about IN activity (100% Ps. syr.-like, higher emissions)

CAM-Oslo, Hoose et al, ERL 2010

Pacific Northwest
NATIONAL LABORATORY

Laboratory investigation of coated/uncoated soot aerosols



Not clear (right of line): immersion mode?

Friedman et al., JGR (2011)

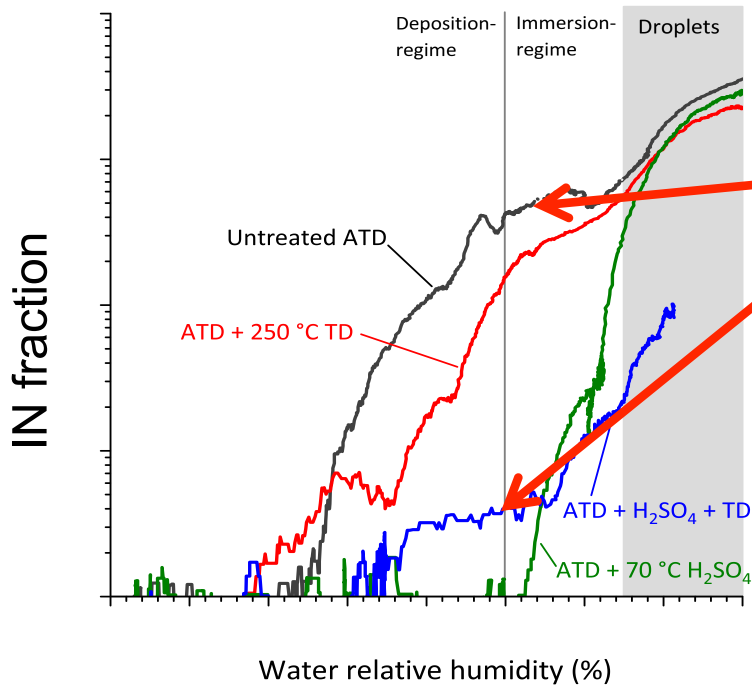
Activated fraction of particles as a function of relative humidity with respect to water (RH_w) and size. Soot particles were coated with adipic acid.

Soot are poor IN in the deposition nucleation mode (left of line), but they can provide the surfaces to condense the water at colder conditions.

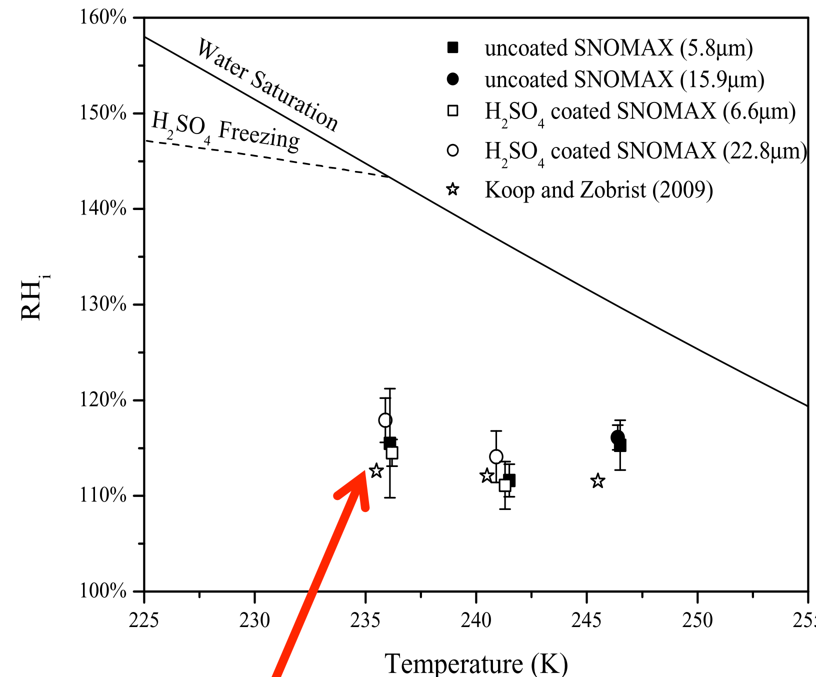
Pacific Northwest
NATIONAL LABORATORY

Understanding the effect of internal mixing (surface coating) on ice nucleation efficiency

deposition immersion



Irreversible loss of ice active sites of ATD due to sulfuric acid coating

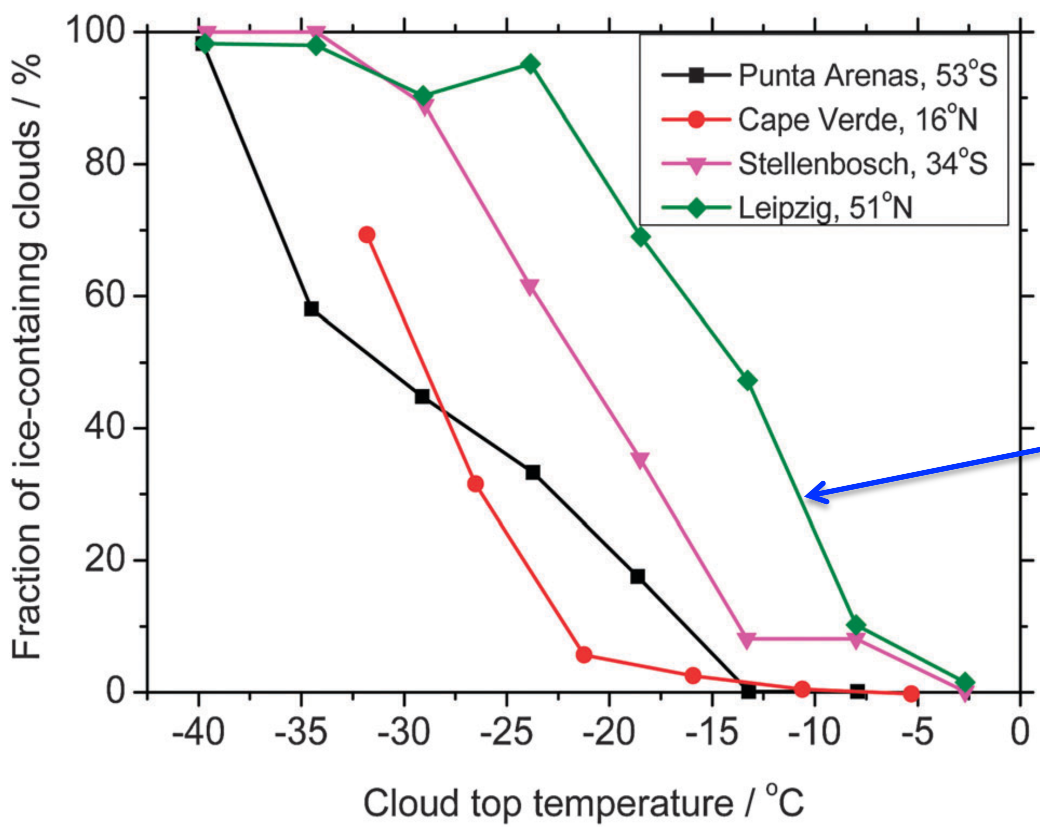


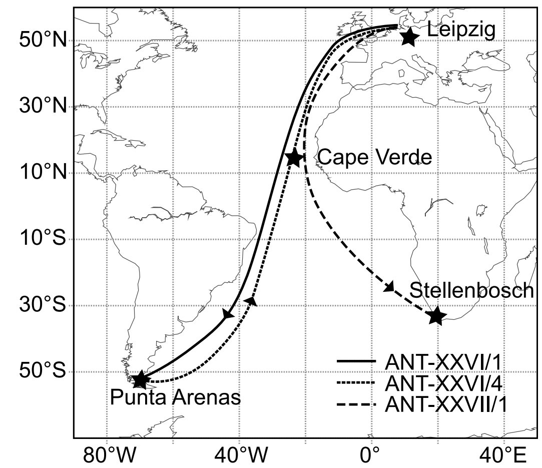
Little effect of sulfate acid coating on SNOMAX (a proxy for biological aerosol)

Sullivan et al., ACP (2010)

Chernoff & Bertram, JGR (2010)

Effect of different types of aerosols on mixed-phase clouds





Anthropogenic IN?

Kanitz et al., GRL (2011)

Pacific Northwest
NATIONAL LABORATORY

Q3. What measurement approaches are needed to characterize all relevant ice nucleation processes?

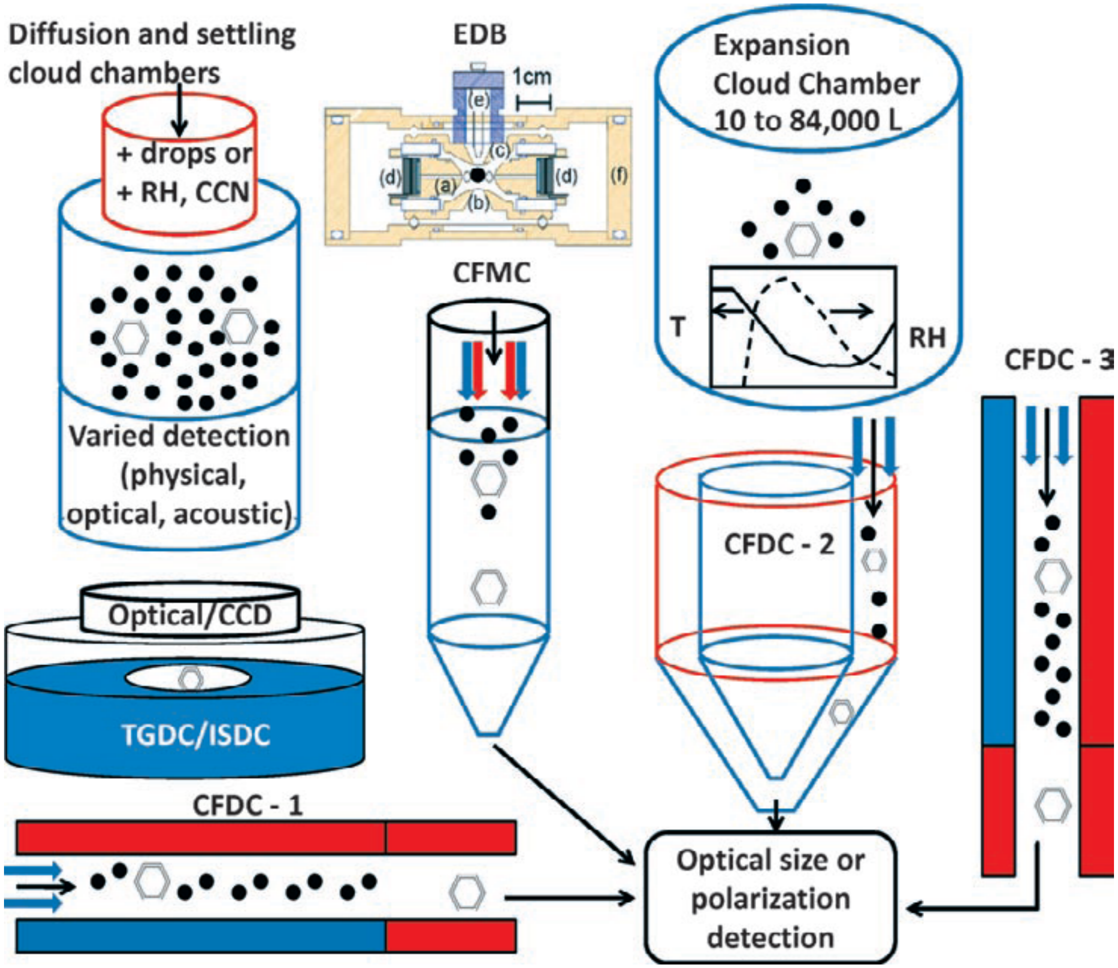
Needs and issues for using different IN measurement methods

- Atmospheric IN activity spans many orders of magnitude (see earlier). Access to the warm temperature regime of very low IN concentrations requires different methods than are easily used at lower temperatures.

  → Need to concentrate air samples or collect large volumes for offline analysis.

- Different methods emphasize different mechanisms, which is needed.
- All have characteristic measurement times and potential artifacts.

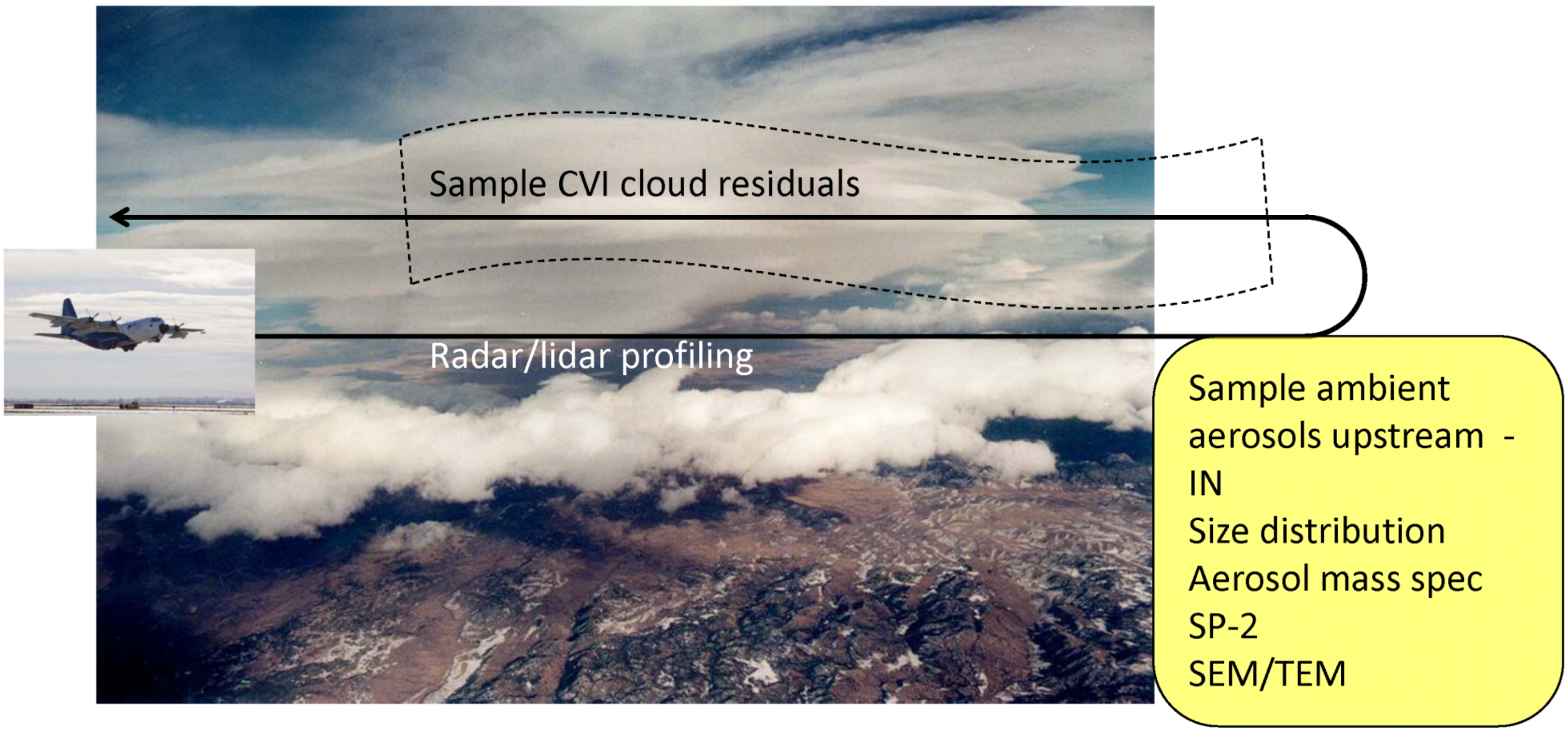
A host of representative IN measurement methods



- -Cloud chambers
- -Continuous flow chambers
- -Particle/droplet traps
- -Static chamber for particle processing or droplet freezing
- -Drop freezing arrays (not shown)

Pacific Northwest NATIONAL LABORATORY

Use clouds to measure IN: e.g., ICE-L methods



Pacific Northwest
NATIONAL LABORATORY

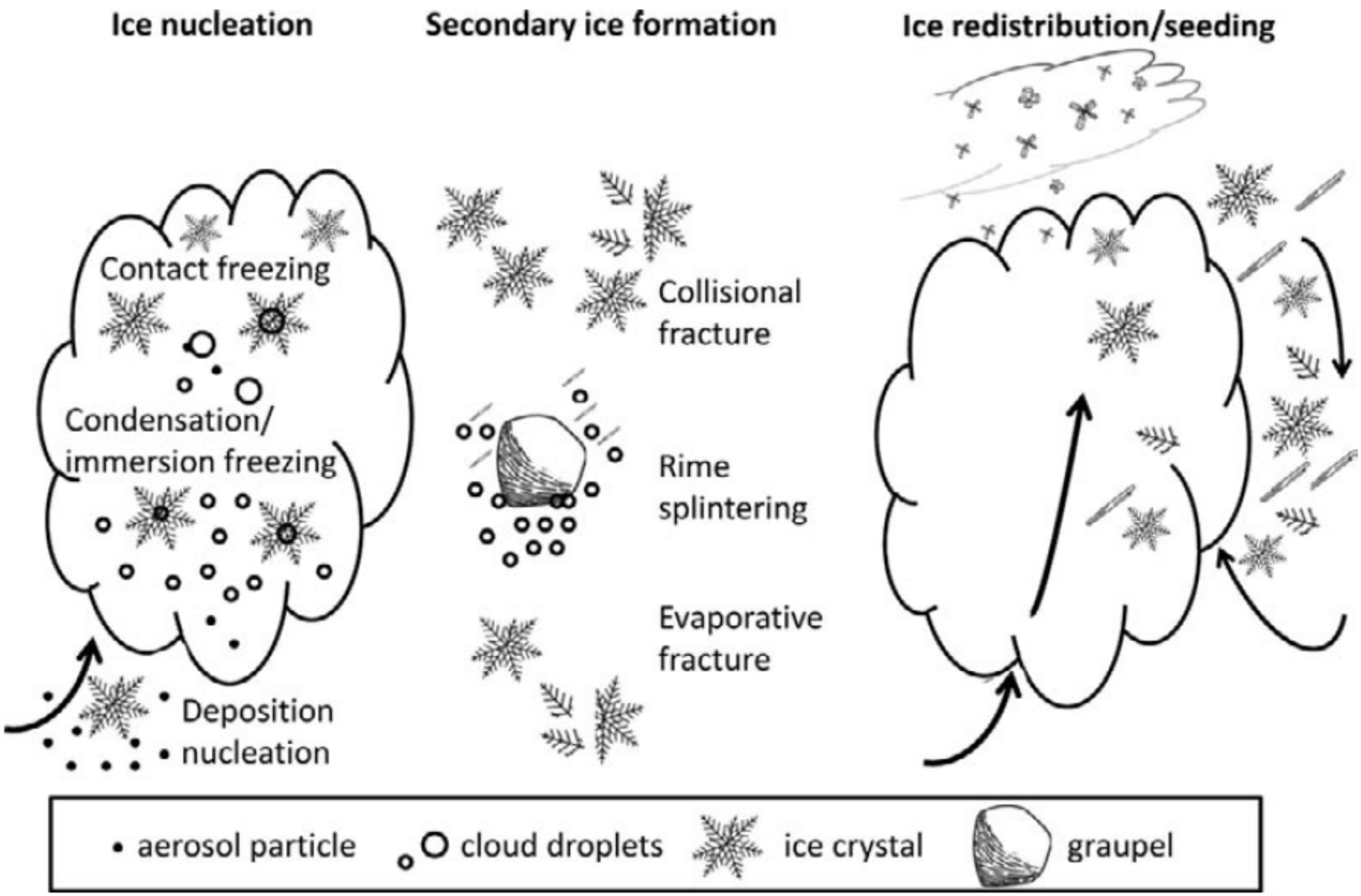
Q4. Can we reach IN – ice crystal closure at current stages of IN and ice crystal measurement? If not, what areas are needed for improvement?

- Inter-consistency between IN and ice crystal numbers in clouds

  - importance of secondary ice formation in some types of clouds

  - role of other cloud processes other than ice nucleation, e.g., aggregation and sedimentation of ice crystals

Ice Nucleation and Ice Lifecycle

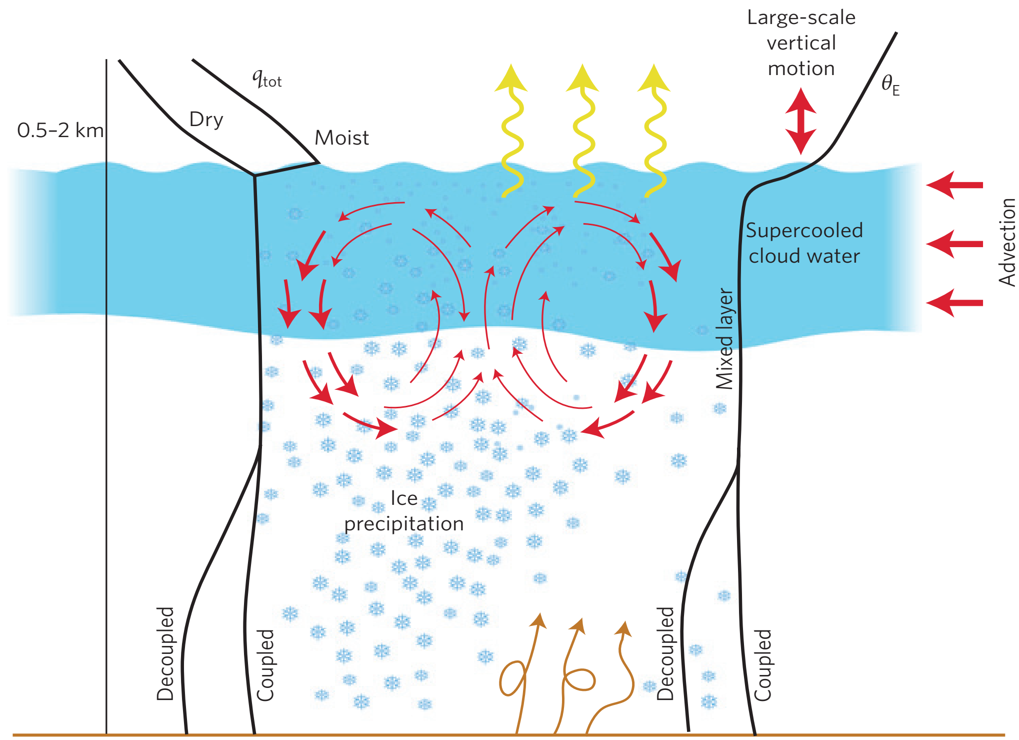


DeMott et al 2011 BAMS

Pacific Northwest
NATIONAL LABORATORY

Q5. What are the roles of ice nucleation on cloud and precipitation properties and climate forcing?

Resilience of persistent Arctic mixed-phase clouds



Radiative Cooling
- Drives buoyant production of turbulence
- Forces direct condensation within inversion layer
- Requires minimum amount of cloud liquid water

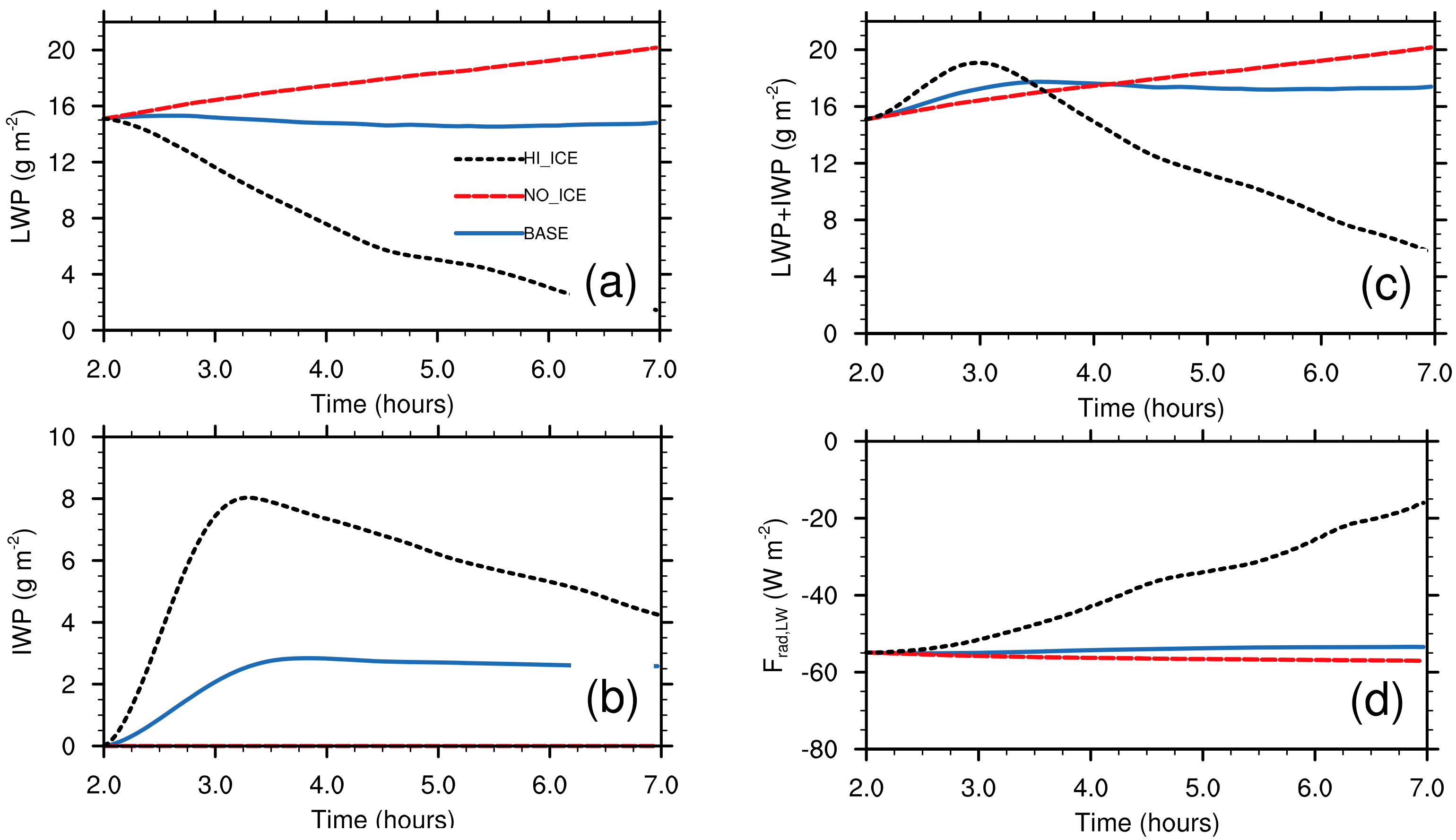
Microphysics
- Liquid forms in updrafts and sometimes within the inversion layer
- Ice nucleates in cloud
- Rapid ice growth promotes sedimentation from cloud

Dynamics
- Cloud-forced turbulent mixed layer with strong narrow downdrafts, weak broad updrafts, and q_{tot} and θ_E nearly constant with height
- Small-scale, weak turbulence in cloudy inversion layer
- Large-scale advection of water vapour important

Surface Layer
- Turbulence and q contributions can be weak or strong
- Sink of atmospheric moisture due to ice precipitation
- Surface type (ocean, ice, land) influences interaction with cloud

Morrison et al. Nature Geo. (2011)

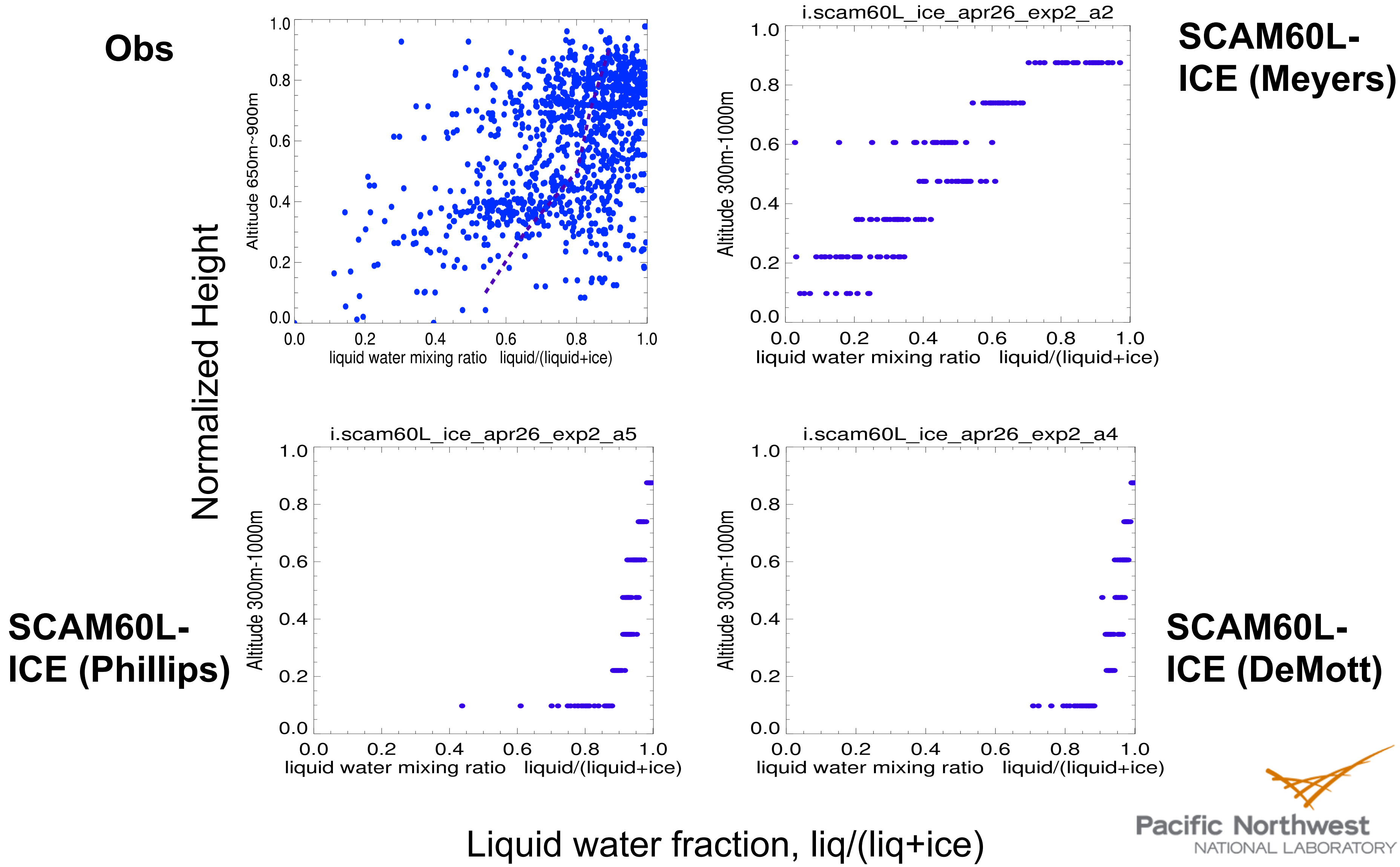
Role of ice crystals on Arctic mixed-phase clouds (ISDAC April 26, 2008)



Ovchinnikov et al. JGR, 2011

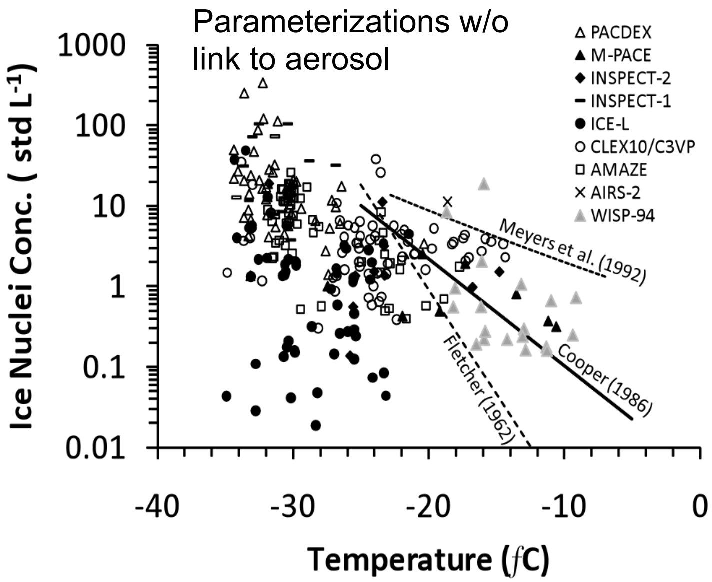
Pacific Northwest NATIONAL LABORATORY

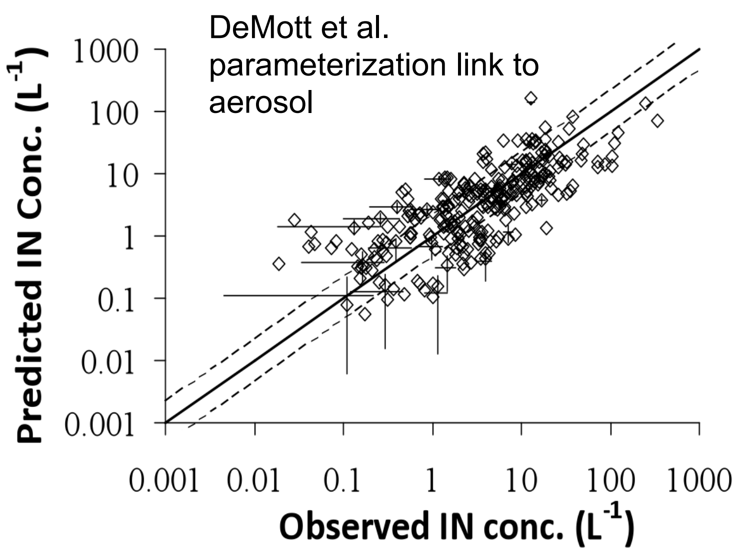
Liquid/ice partitioning in single-layer mixed-phase clouds during ISDAC (April 26)



Effect of ice nucleation in mixed-phase clouds

Parameterized vs. observed IN

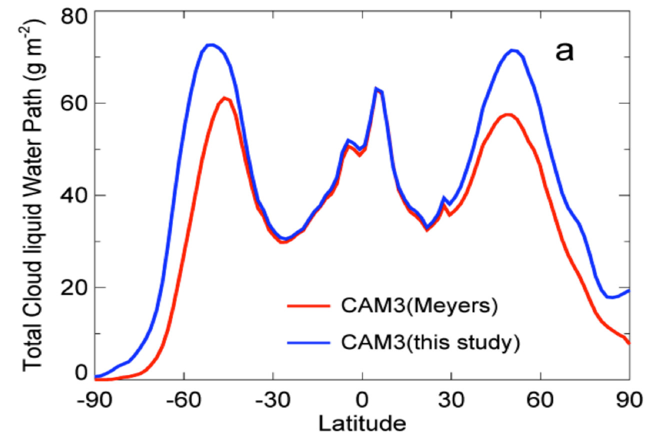


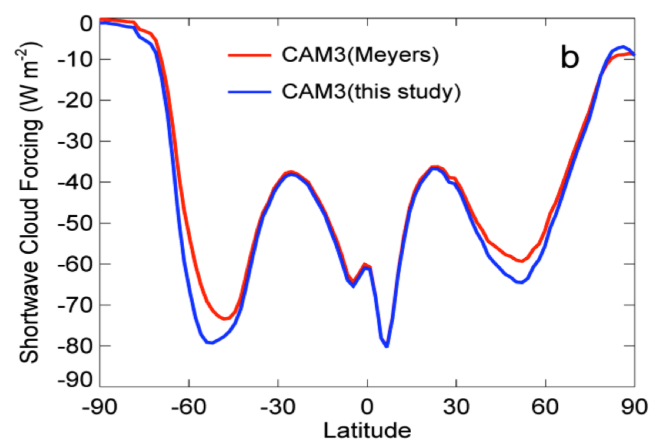


Multiple ice nucleation mechanisms related to aerosol & meteorological conditions

Uncertainties in IN parameterizations produce significant changes in CAM modeled LWP, cloud forcing and cloud fraction

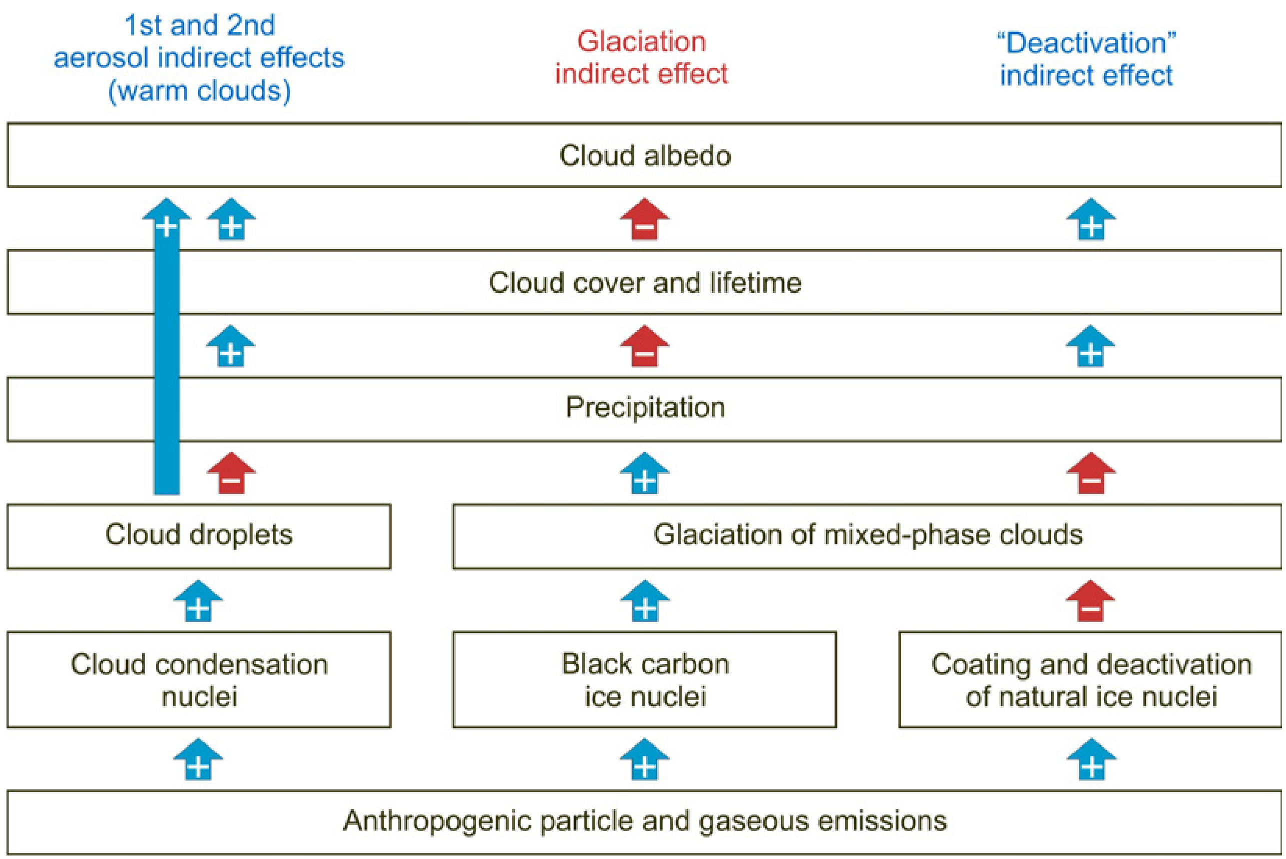
LWP and SWCF with two IN parameterizations





DeMott et al. (2010)

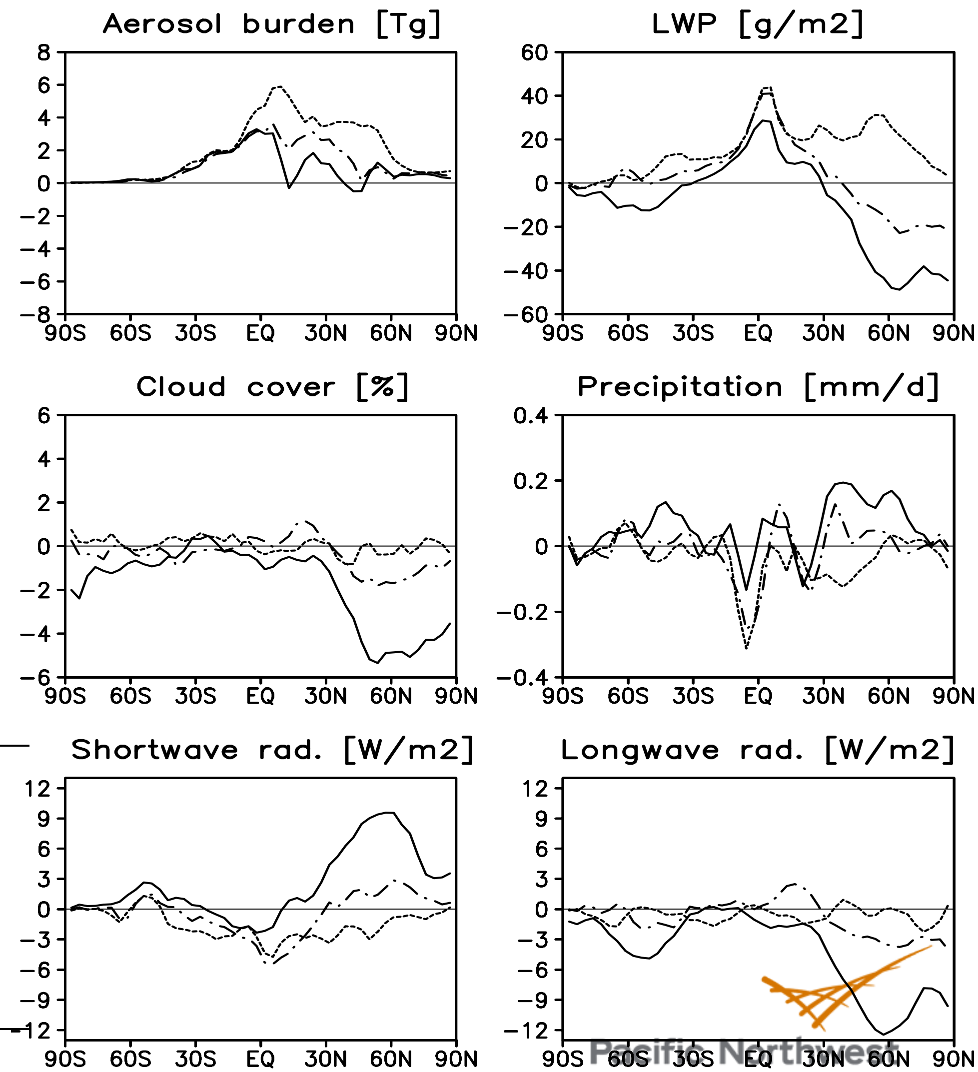
Aerosol indirect effect in warm and mixed-phase clouds



Lohmann GRL (2002); Hoose et al. ERL (2008)

Aerosol indirect effect in warm and mixed-phase clouds

- If BC were active as IN, enhanced glaciation would deplete LWP and cloud cover
- Impacts on SW and LW cloud forcing largely cancel
- BC is a poor IN, but other anthropogenic mechanisms are possible



Simulation	BC10%	BC1%	BC0%
ΔAB	1.1 (9)	1.6 (13)	2.6 (22)
ΔLWP g m^{-2}	−2.9 (−3)	7.9 (8)	18.5 (18)
ΔIWP g m^{-2}	0.6 (8)	0.2 (3)	0.1 (1)
ΔTCC	−1.3 (−2)	−0.3 (0)	0. (0)
ΔPR mm d^{-1}	0.05 (2)	−0.02 (−1)	−0.05 (−2)
ΔF_{SW} W m^{-2}	+1.9 (1)	−1.0 (0)	−2.0 (−1)
ΔF_{LW} W m^{-2}	−3.5 (2)	−0.5 (0)	−0.6 (−0)
ΔF_{net} W m^{-2}	−1.6	−1.5	−1.4

Lohmann GRL (2002)

A Roadmap

- Intercompare existing instruments with different designs and/or operational principles.
- Explore consistency in interpretation of ice nucleation data
- Define capabilities/uncertainties for measuring atmospheric IN concentrations (versus "high signal" lab data)

→Organize a workshop in the laboratory and field (as in DeMott et al. 2011)

Pacific Northwest
NATIONAL LABORATORY

A Roadmap

- Laboratory investigation of ice nucleation mechanisms at process level.

  - soot in the immersion mode
  - effect of surface coating, including coating thickness
  - ice nucleation at cold (T<-60C) and warm (T>-15C) temperatures
  - role of biological aerosol (e.g., from marine source)
  - contact freezing

Pacific Northwest
NATIONAL LABORATORY

A Roadmap

- Propose dedicated IOPs on ice nuclei in a specific aerosol source region (desert, industrial pollution, fire, biological aerosol, and soot).
- Explore other opportunities for ice nuclei measurements in different locations (e.g., GOAmazon 2014+1); routine IN measurements at ACRF sites.
- Propose an ACRF measurement in Southern Ocean would be a good place to study glaciation of mixed-phase clouds in SH storm track region.

Pacific Northwest
NATIONAL LABORATORY

A Roadmap

- Analysis of current data : ISDAC, M-PACE, SpartiCus, MC3E
  Ni, IWC – T, RH, w, aerosol (size, composition)

- Process level understanding with cloud resolving models (CRM) and single column model (SCM) for IOP and ACRF cases
  - examine the roles of ice nucleation and other cloud processes on ice-containing clouds (Arctic stratus, cirrus and convective clouds)

- Quantify the uncertainties of ice nucleation on ice-containing clouds and radiative forcing in climate models

Pacific Northwest
NATIONAL LABORATORY