Do biomass burning particles nucleate ice?

[courtesy of Hans Moosmueller]

Biomass burning smoke are a large point source of aerosol

[www.atmos.washington.edu]

When smoke is entrained into clouds the particles function as cloud condensation and **perhaps ice nuclei**

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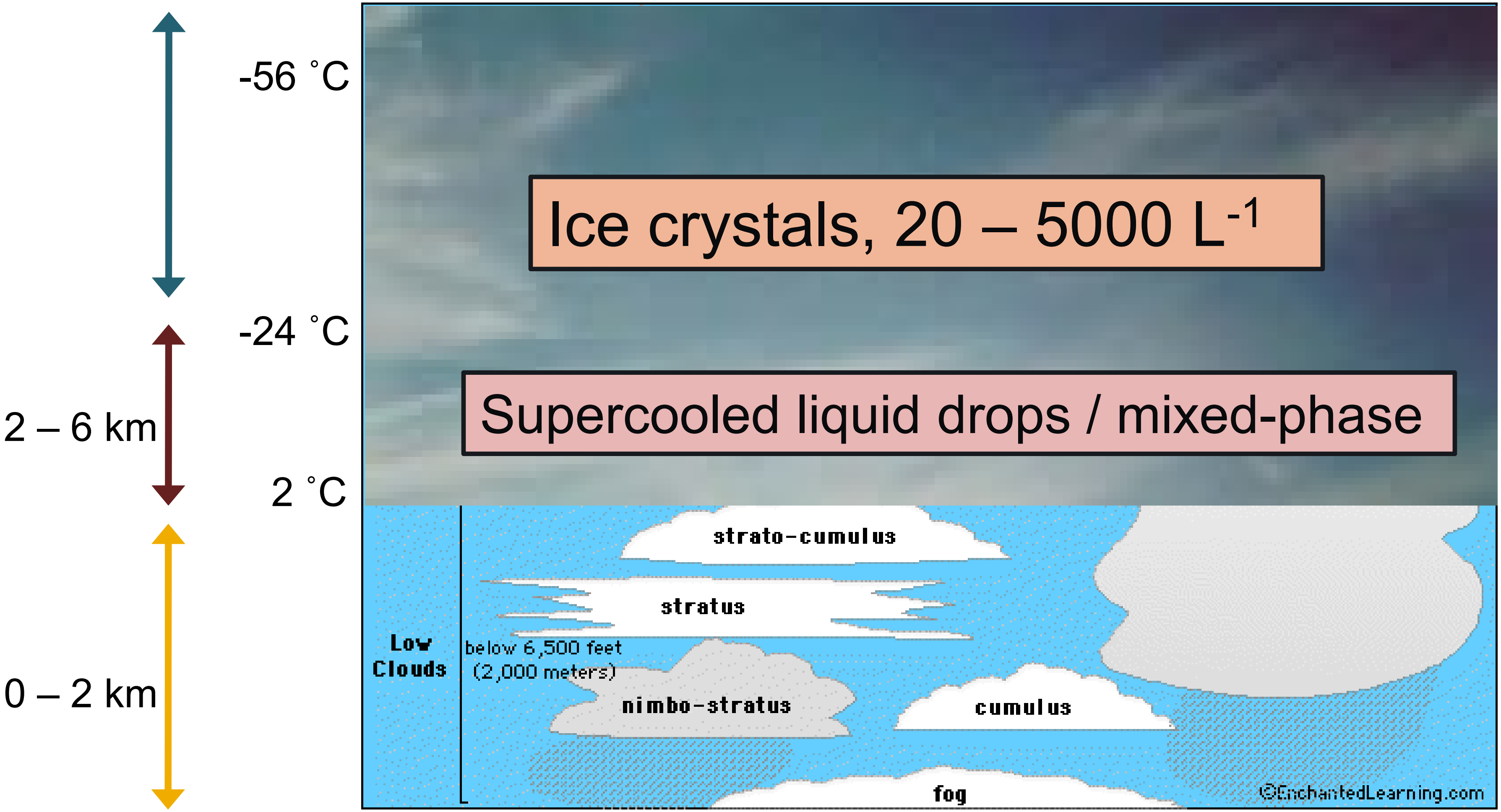
Collaborators

- Mathews Parsons, Kip Carrico, Anthony Prenni, Paul DeMott, Sonia Kreidenweis
- Amy Sullivan (URG data, OC/EC chemical analysis)
- Gavin McMeeking (MCE and gas phase emission factors)
- Wei Min Hao, Jeff Collett, Hans Moosmüller, William Malm
- Cyle Wold and all the members of the FSL facility in Missoula

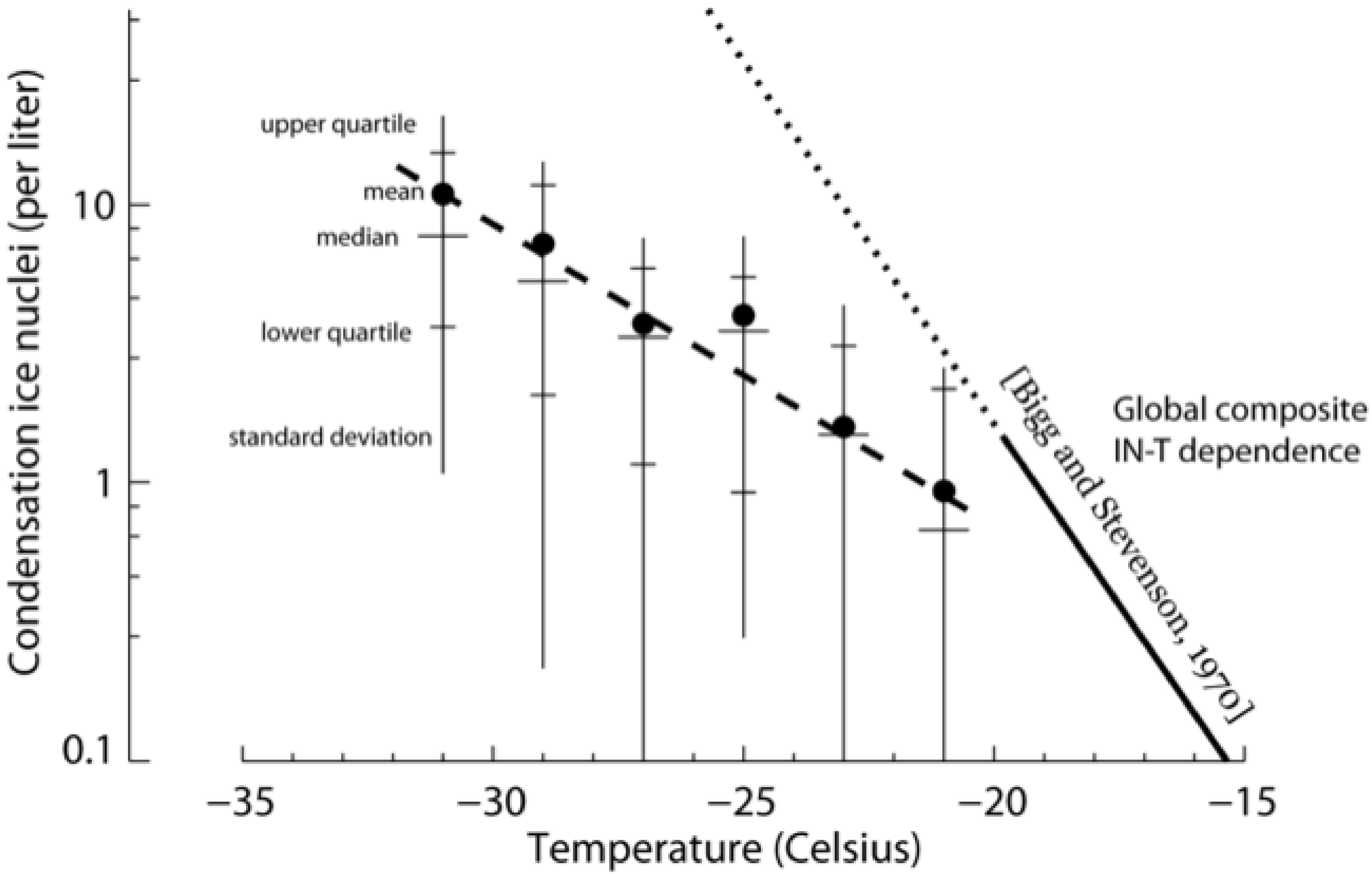
- EPA (FLAME-II main study)
- NASA ROSES A.7 (Freezing studies)

Cold clouds – ice crystals often initiate precipitation through the Bergeron process



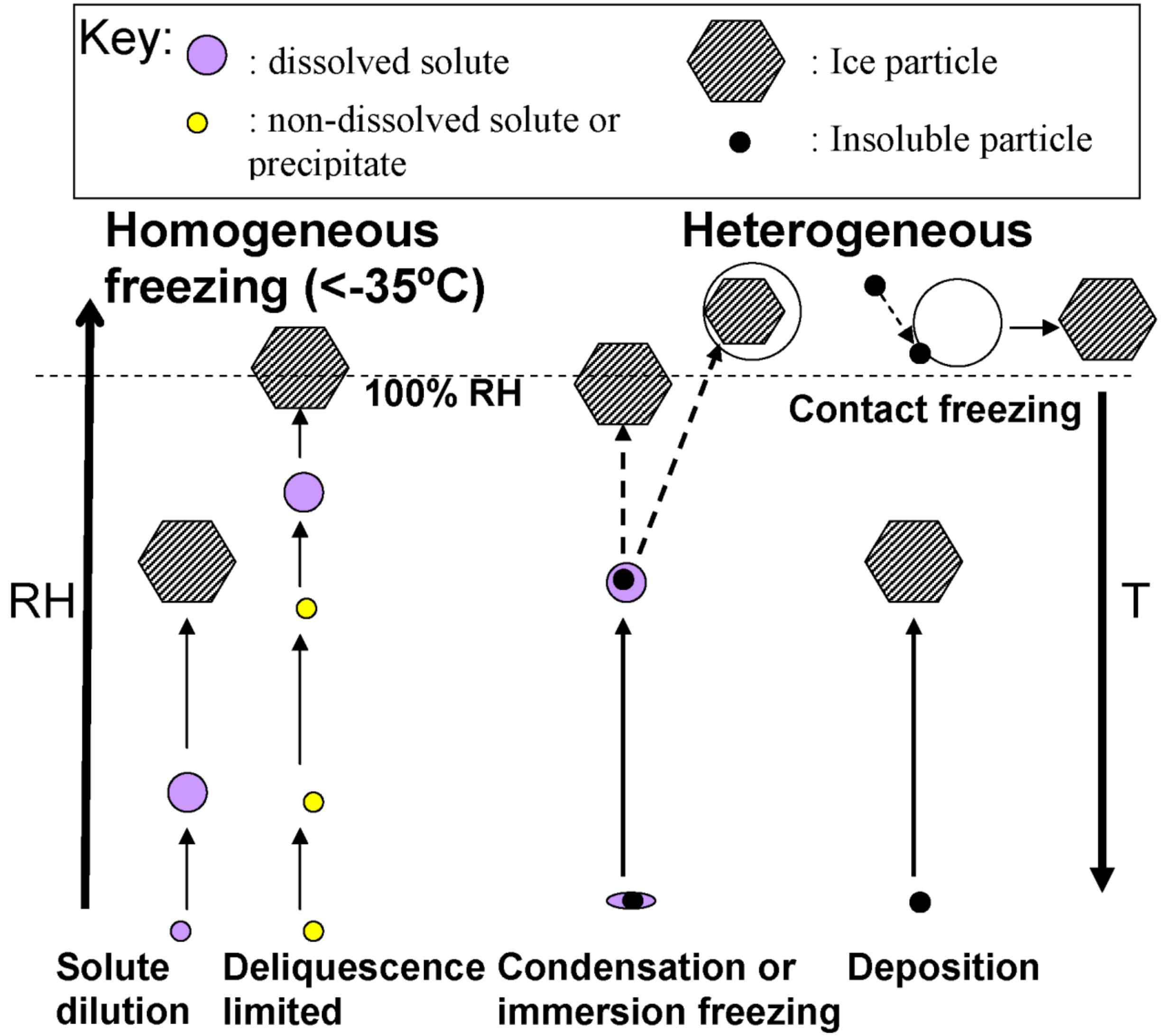
http://www.enchantedlearning.com/subjects/astronomy/planets/earth/clouds/

In the atmosphere, ice nuclei are rare (1:10^5 particles)

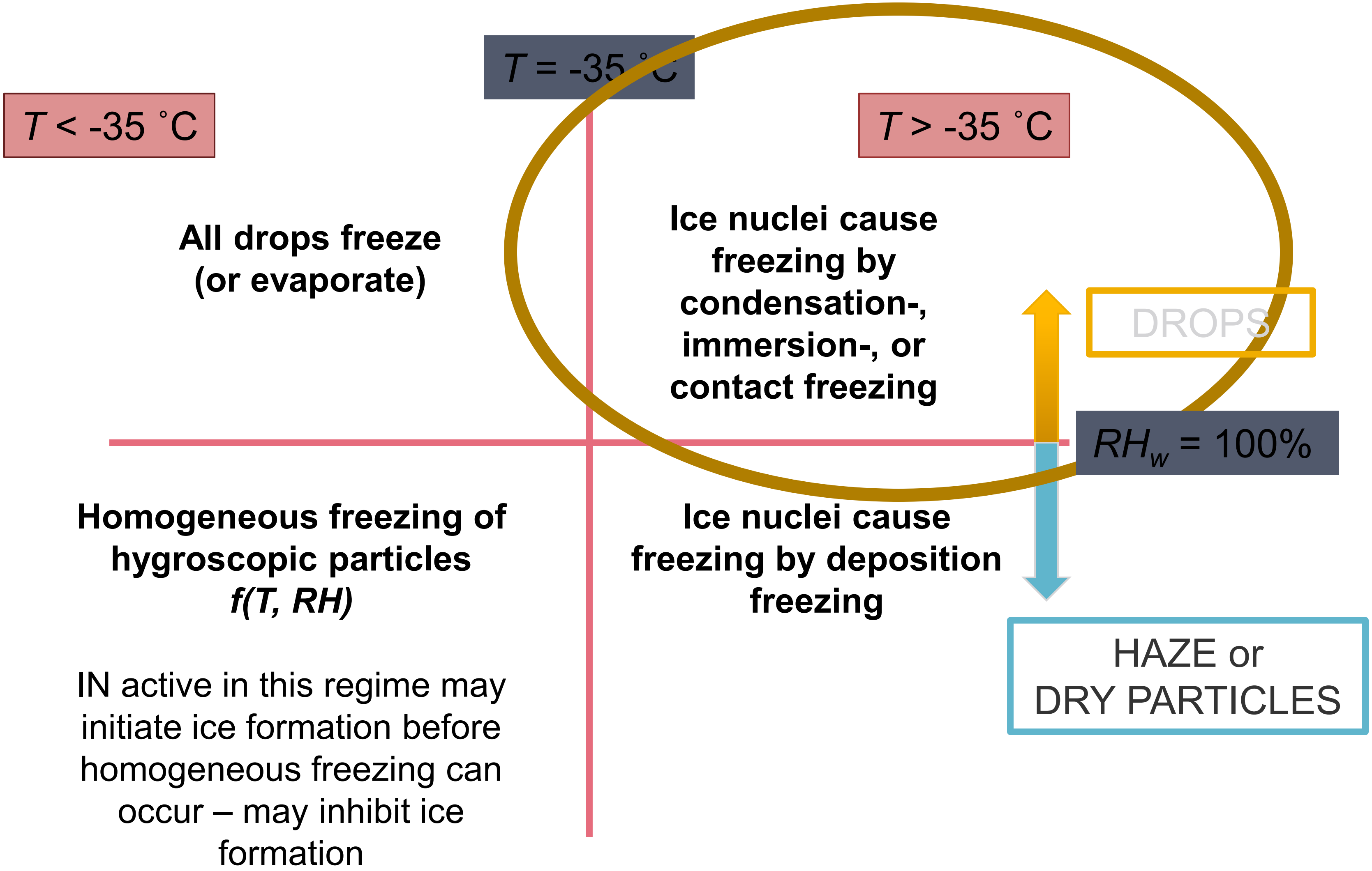


Amazon, wet season, Prenni et al., 2009

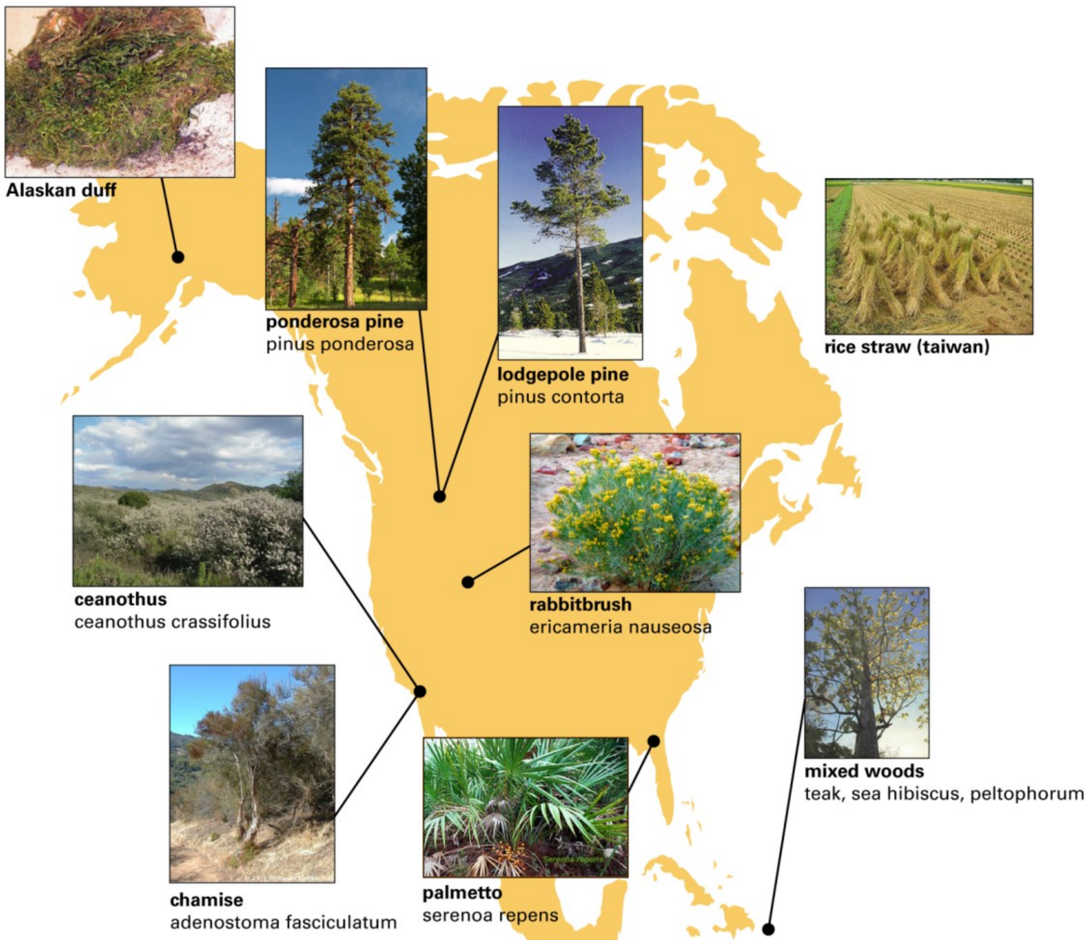
Aerosols nucleate ice by varied mechanisms, most depending at least on T, some on RH, chemistry, and aerosol mechanics.



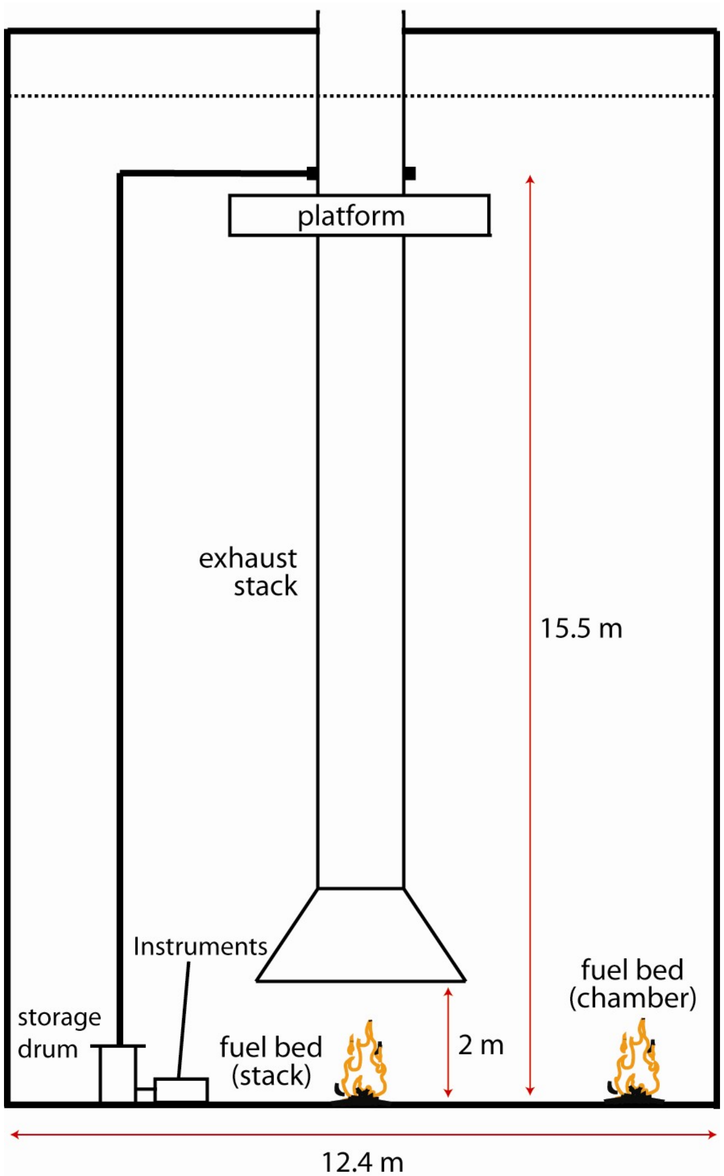
Ice nucleation mechanisms, another view



Fire Lab at Missoula Experiment (FLAME-II): controlled burns of ~30 fuels from North America

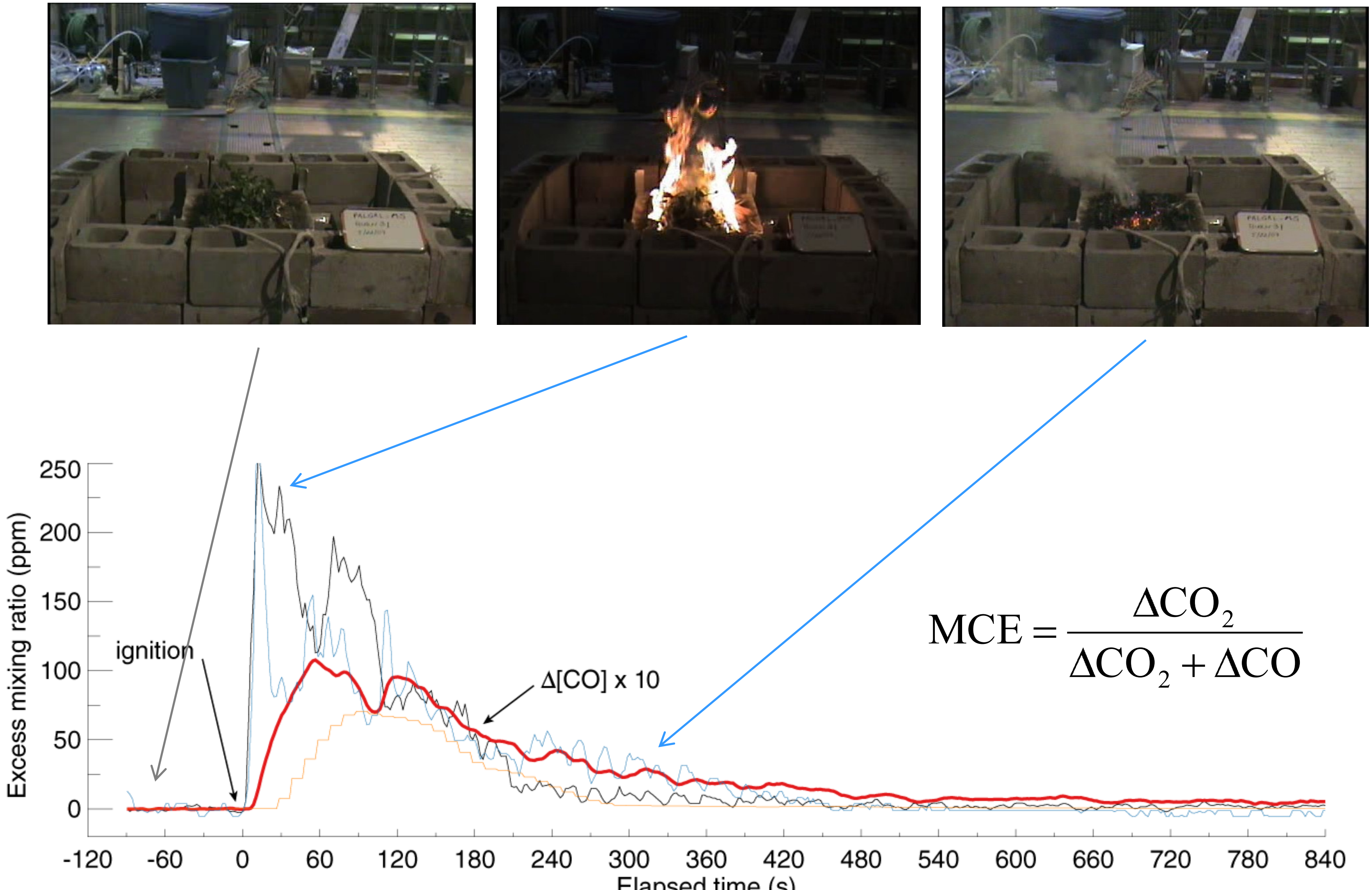


plant images courtesy santa monica mountains trails council, bay area hicker, alberta parks and recreation, daniel kirk, food and agriculture organization of the United Nations



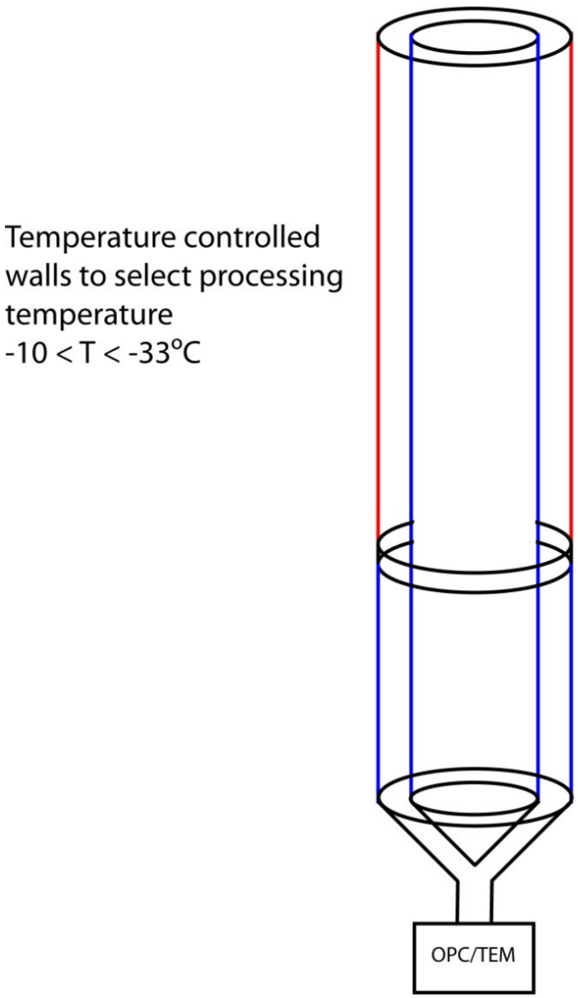
Trace gas emissions/burn example

INITIAL **FLAMING** **SMOLDERING**

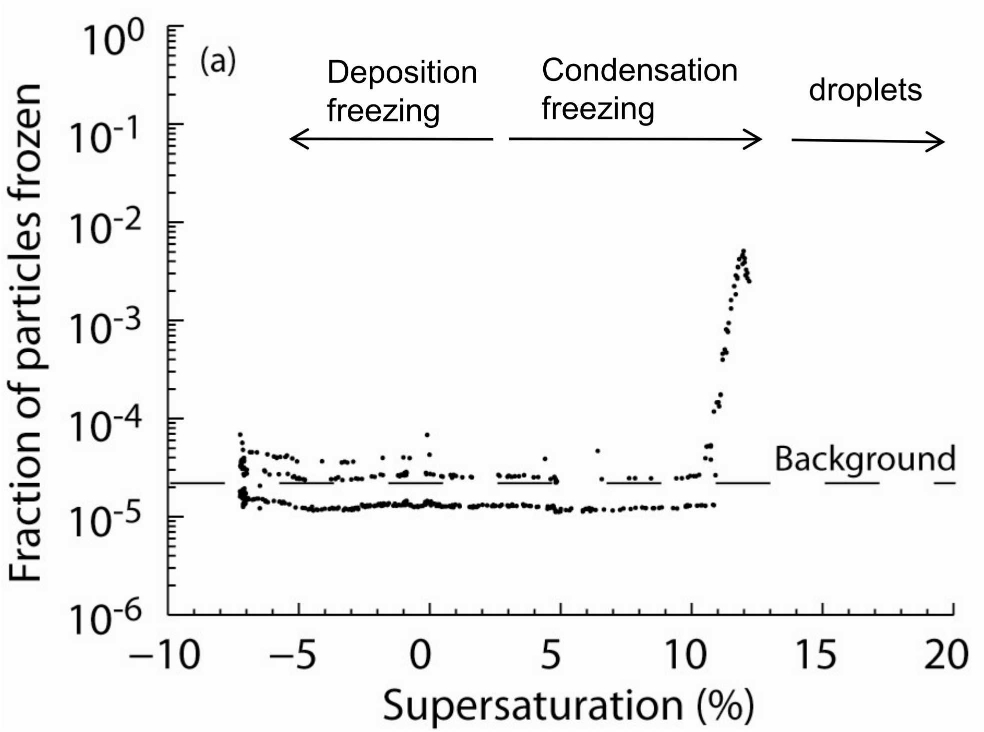


$$\mathrm{MCE} = \frac{\Delta CO_2}{\Delta CO_2 + \Delta CO}$$

Ice nuclei were measured using the Colorado State University continuous flow diffusion counter (CFDC)



Measurements of ice nuclei using the CSU Continuous Flow Diffusion Chamber (CFDC)



Ammonium sulfate, T = -30C

Detection limit (noise) ~1:100,000

Evaporation region good to ~11% water supersaturation (droplets persist at higher SS_w)

Define ice nucleation efficiency parameter as maximum fraction frozen

- Condensation/immersion ice nuclei at -30˚C
- Polydisperse aerosol (D < 1.5μm, impactor)

$\xi = \log_{10}$ maximum activated fraction

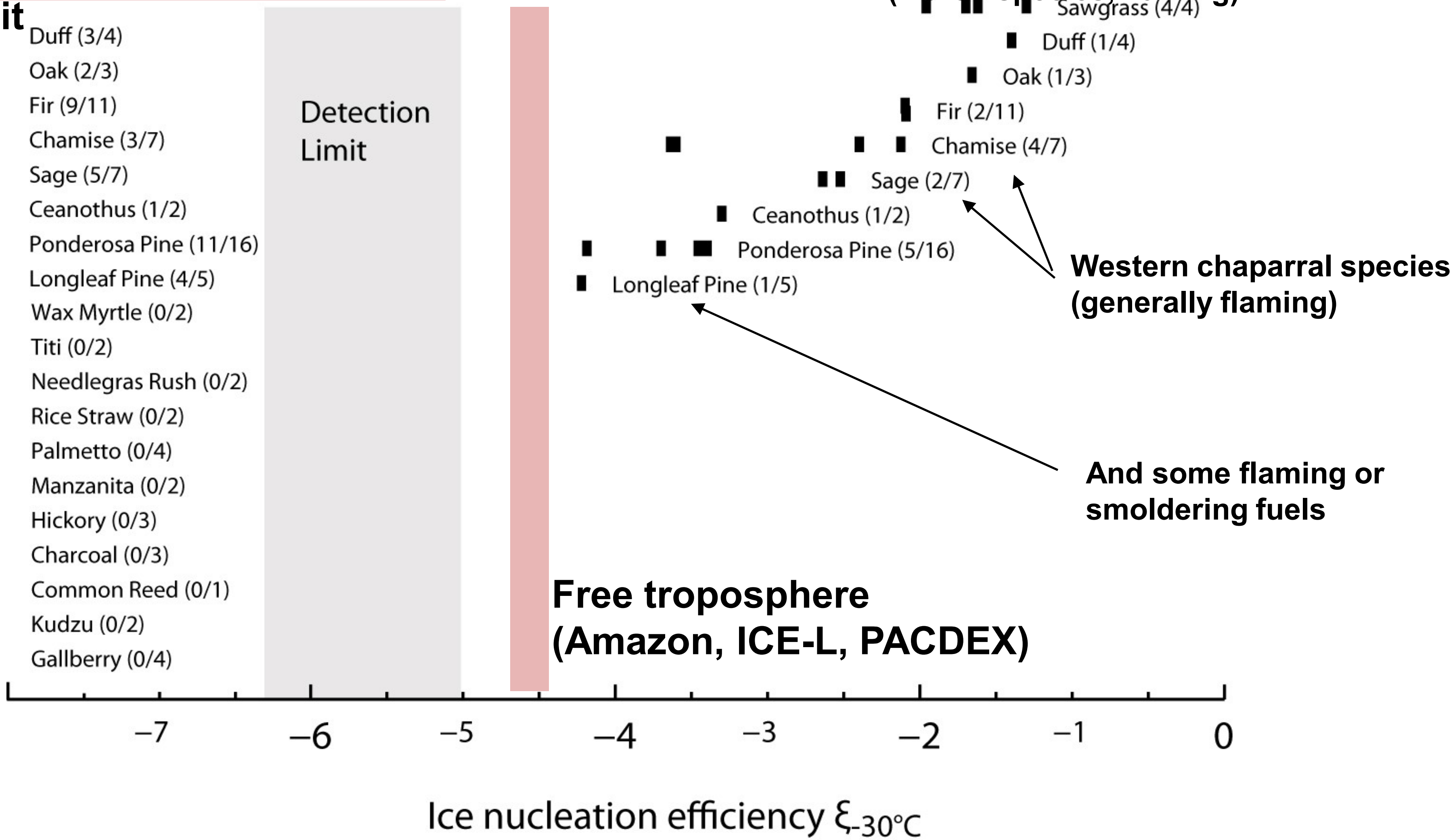
IN efficiency generally decreases at warmer temperatures

$\xi_{-30^\circ C}$ is an upper estimate of potential IN emissions into the atmosphere (most mixed-phase clouds are warmer).

Some fuels seem to preferentially produce ice nuclei but not all the time

80% no ice nucleation signal above detection limit

There is something about sawgrass (marsh species, flaming)



The ice nucleation efficiency can directly be used in fire emission inventories

Modifi

Crude estimate of impact:

1. Take area of boreal forest burned/year
2. Assume typical pine tree emission factor
3. Assume ~1 week lifetime for aerosol
4. Assume vertical extent of plume
5. Compare against typical IN background concentrations

~ 4000x4000 km^2 region impacted

Ice nuclei
emission

the fuel
burned

mass ratio

$$EF = 5 \;\; 10^{6} - 3.4 \;\; 10^{15} \text{ IN m}^{-2}$$

Fire and ice: potential mechanisms for biomass burning to produce IN

Insoluble salts	**Specific organics**	**Metal oxides**	**Soot or EC**
halides (AgI) *specific crystal structure* *crystal defects*	*long chain alcohols, aromatics, amino acids* *(need to be dissolved in H_2O)*	*mineral dust* *insoluble active sites*	*active sites* *mesopore water*

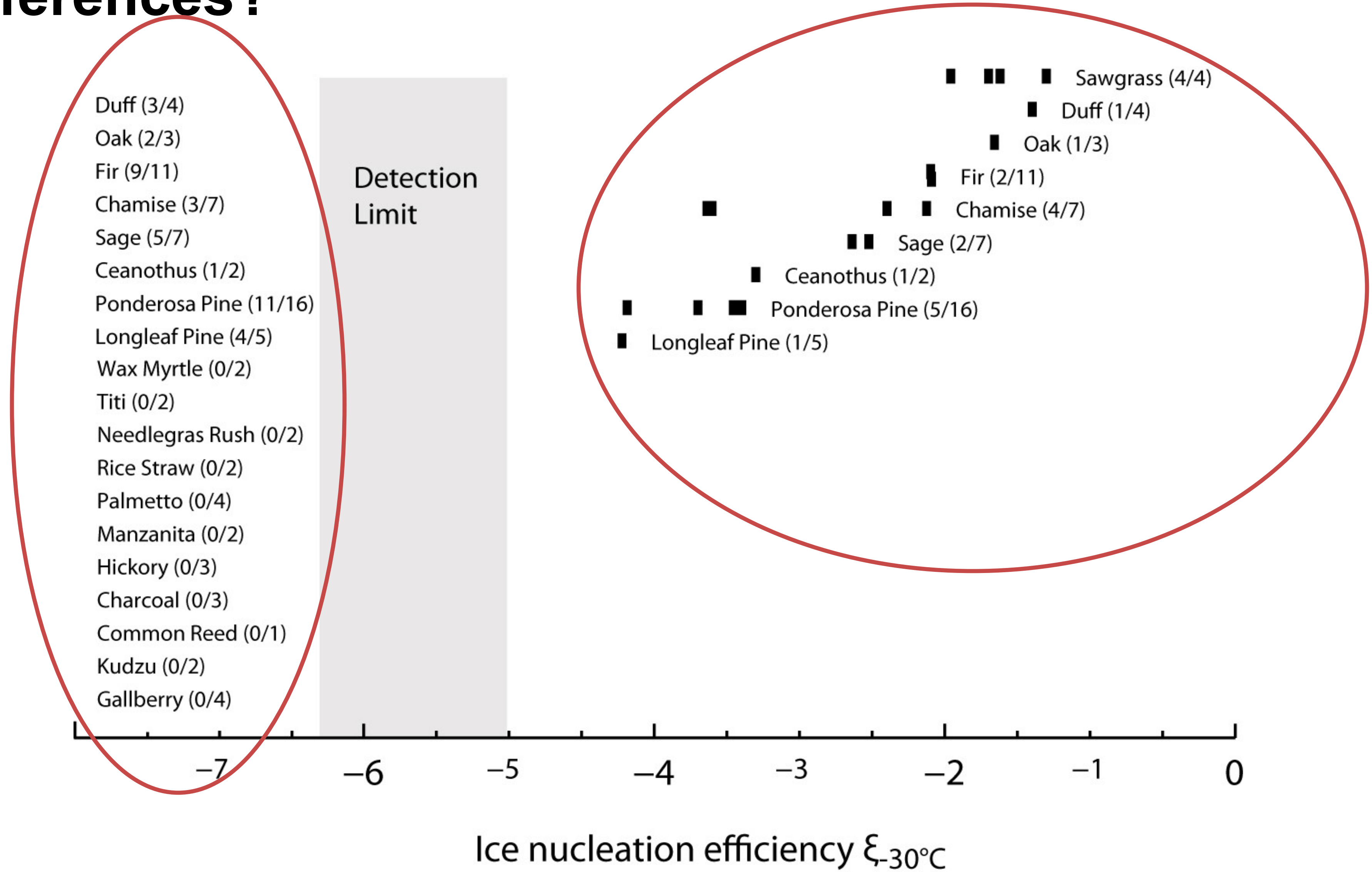
Secondary processes
- partitioning of semi-volatile organics
- heterogeneous reactions with surfaces
- particle phase chemistry (e.g. oligomerization)

Emissions
- aerosol number, surface area, mass
- aerosol composition, hygroscopiciy, mixing state

Combustion conditions
- combustion efficiency
- fire radiative power
- fire temperature

Fuel type, fuel moisture
Ash fraction, composition
Soil composition (may be lofted with fire)

Question: what is difference in smoke composition or combustion conditions that explain these differences?



Use statistical test to derive probability that the mean properties differ

1. Pool sample population into two groups. Identify any parameter that may be significant (composition, combustion conditions, fuel moisture, etc.) and calculate probability (**P**) that the means are different

Group A $\xi_{-30^{\circ}C}$ < detection limit (~-6)	Group B $\xi_{-30^{\circ}C}$ > detection limit
X samples	Y samples
$mean_A$ (parameter)	$mean_B$ (parameter)

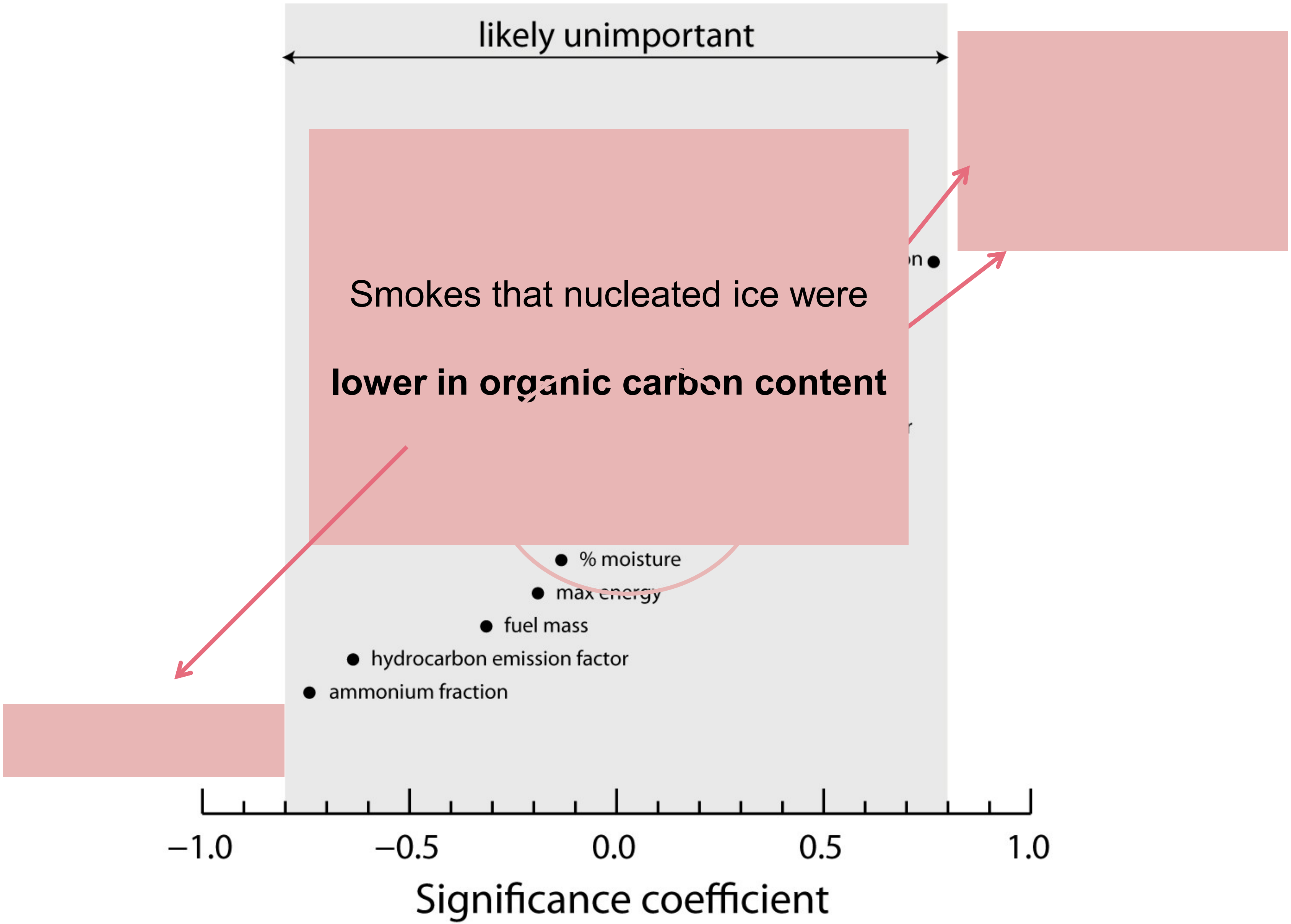
2. Define significance coefficient (S) as

$$S = -.87$$

$$S = \mathrm{gn}(\mu - \iota)P$$

Interpretation: There is an 87% probability that average mean of X was smaller when generating smokes that produced ice nuclei

Summary of significance coefficients



Summary

1. Approximately 20% of samples emitted ice nuclei

2. IN emissions are tied to the fuel type to some degree

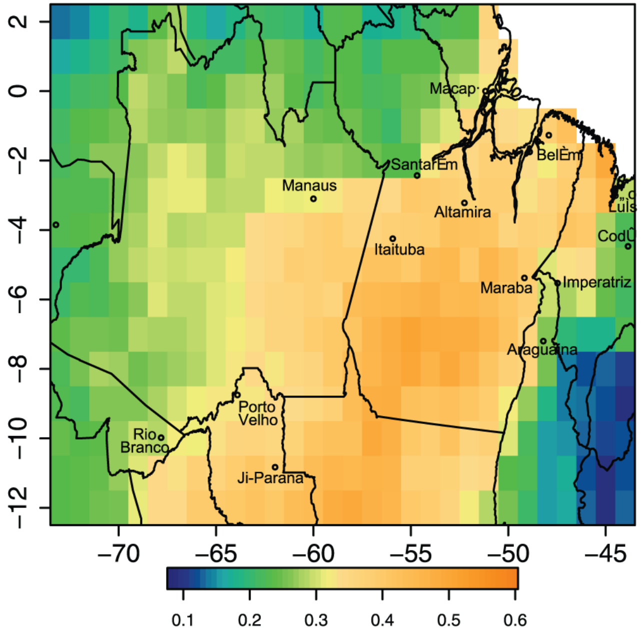
3. Estimates of emission factors suggest regional scale disturbance of IN budget

4. Necessary conditions for IN emissions:
   - High MCE/flaming combustion phase
   - Presence of water soluble inorganic ions
   - Low organic carbon fractions

5. Seemingly unrelated to black carbon/soot

Satellite data suggest that emission of ice nuclei from biomass burning have a regional influence

Spatial Distribution of Average MODIS Aerosol Optical Depth (Aug-Oct 2000)



[Figure adapted from Lin et al., 2006]

Fuels (Pictures courtesy of Hans Moosmueller)

Excelsior (Poplar Product)

Montana Grass

Ponderosa Pine Wood Sticks

Ponderosa Pine Needles

White Pine Needles

Zambia Grass

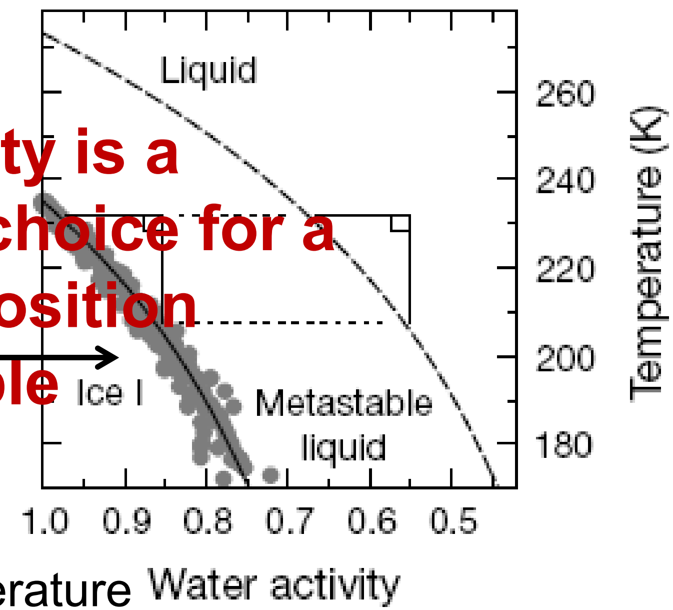
Sage Brush

Tundra Core

Observations suggest that homogeneous nucleation depends on water activity

data collapse on Δa_w which is the basis of parameterization

droplet must dilute to a critical water activity before it freezes



[Koop et al., 2000]

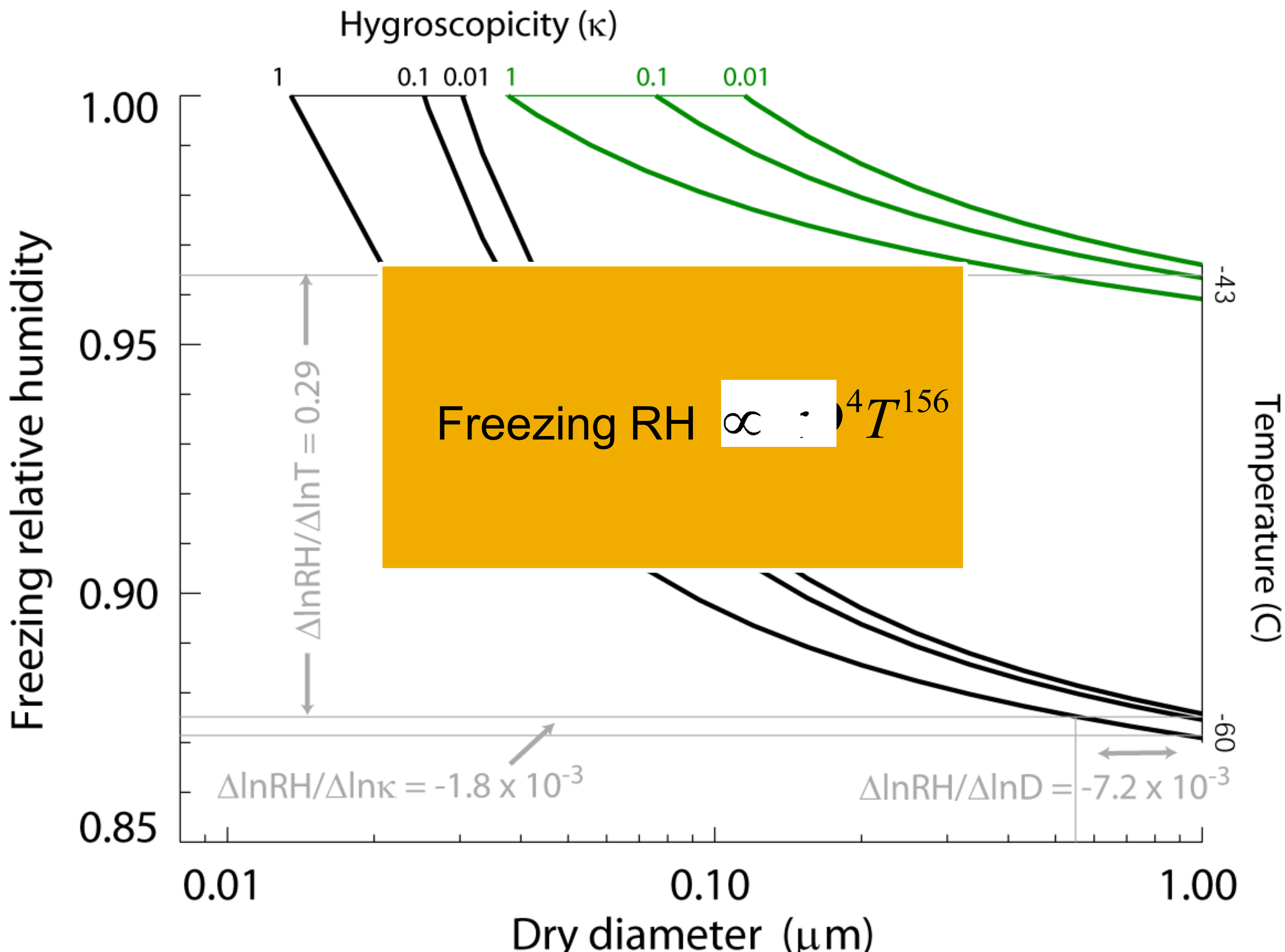
Physical parameters of Koop et al. parameterization (4-dimensional problem)

- Nucleation rates are parameterized based on Δwater activity and droplet volume
- Temperature dependence based on freezing point depression
- Hygroscopicity (κ) relates droplet volume to effective water activity
- Kelvin effect introduces size dependence into freezing

Freezing thus depends in principle on particle

temperature, relative humidity, size, and composition

The relationship between temperature, relative humidity, size, and composition for freezing



[Kreidenweis et al., 2008]

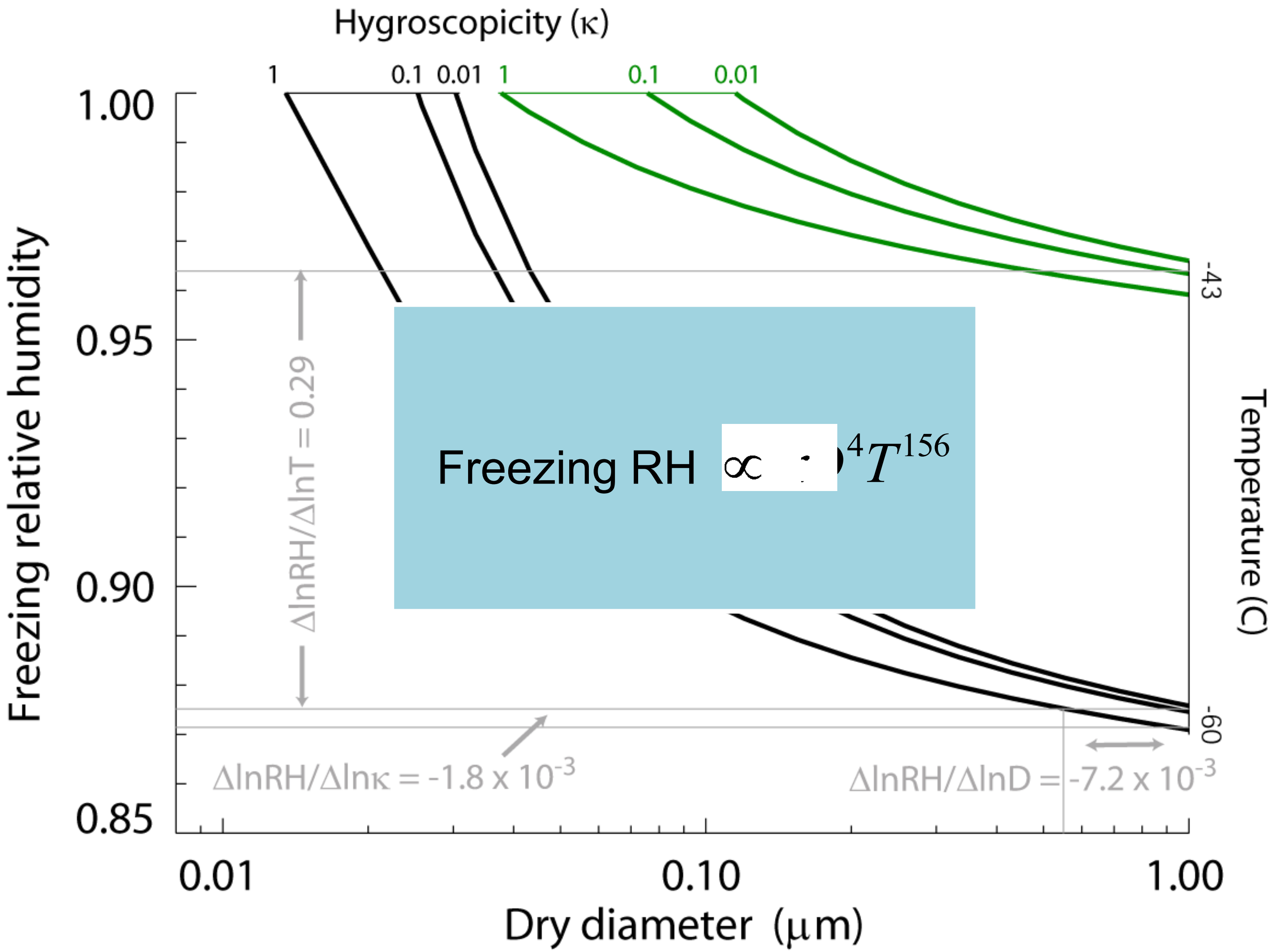
Each of the processes we looked at is more sensitive to diameter than composition (κ)

Wet scattering	Cloud activation	Homogeneous freezing
κD^2 to κD^6	κD^3	$\kappa D^4 T^{156}$

In the atmosphere hygroscopcity and diameter are not independent. Processes that modify composition often also affect particle size.

- Condensation
- Coagulation
- Chemical reactions
- Emissions

The relationship between temperature, relative humidity, size, and composition for freezing



Freezing RH $\propto$ … $^4T^{156}$

[Kreidenweis et al., 2008]