Nucleation to cubic ice

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Cubic ice (Ic)

- Normal ice is hexagonal **(Ih).**
- Cubic ice has same bonds, different packing.
- Distinguishing **Ih** and **Ic** requires X-ray or electron diffraction.
- Cubic ice is metastable.

  => Higher vapor pressure than **Ih**
- Most other properties are nearly identical for **Ic** and **Ih.**

Formation of cubic ice

All available experiments show that water below ~200 K initially freezes to Ic.

+ vapor deposition to surfaces *(many authors)*

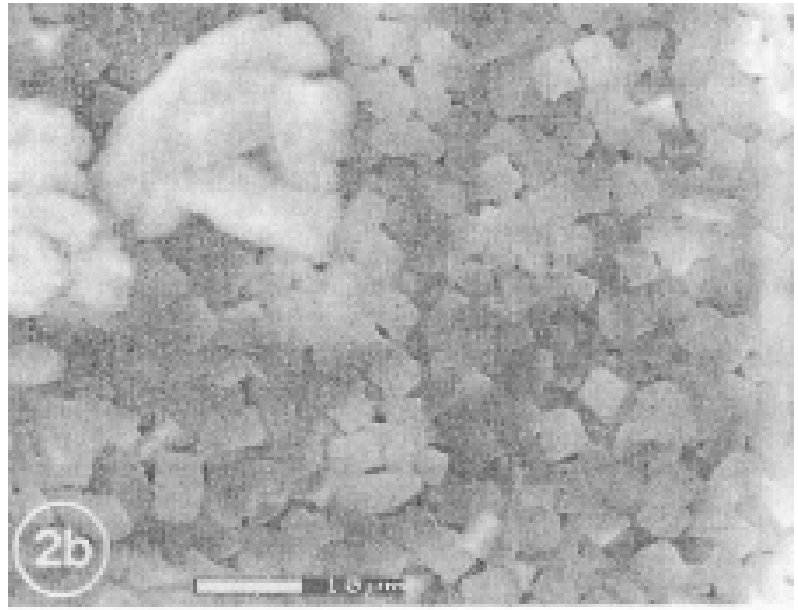
+ liquid droplets hitting cryoplate *(Mayer and Hallbrucker)*

+ nano-droplets in a supersonic jet expansion *(Huang and Bartlett)*

(None of these techniques is how water freezes in the atmosphere.)

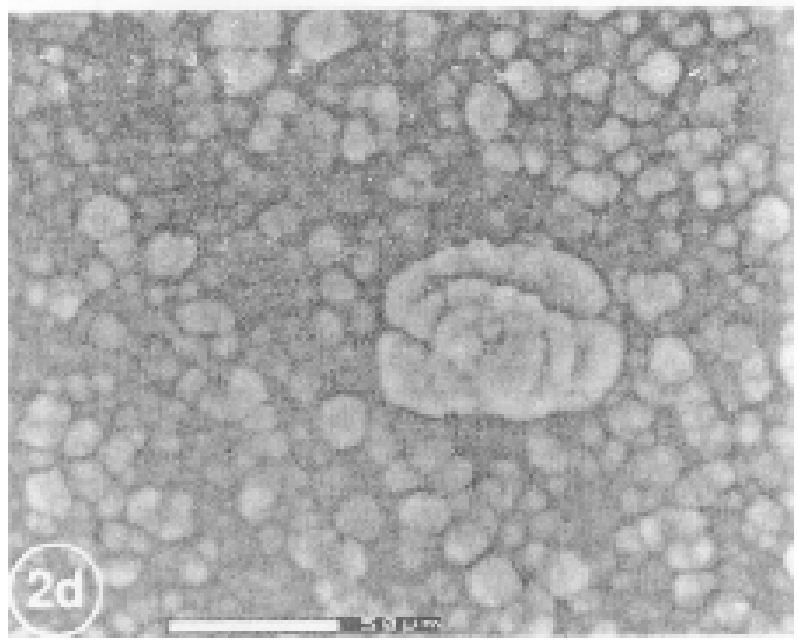
Ice annealing

Ic

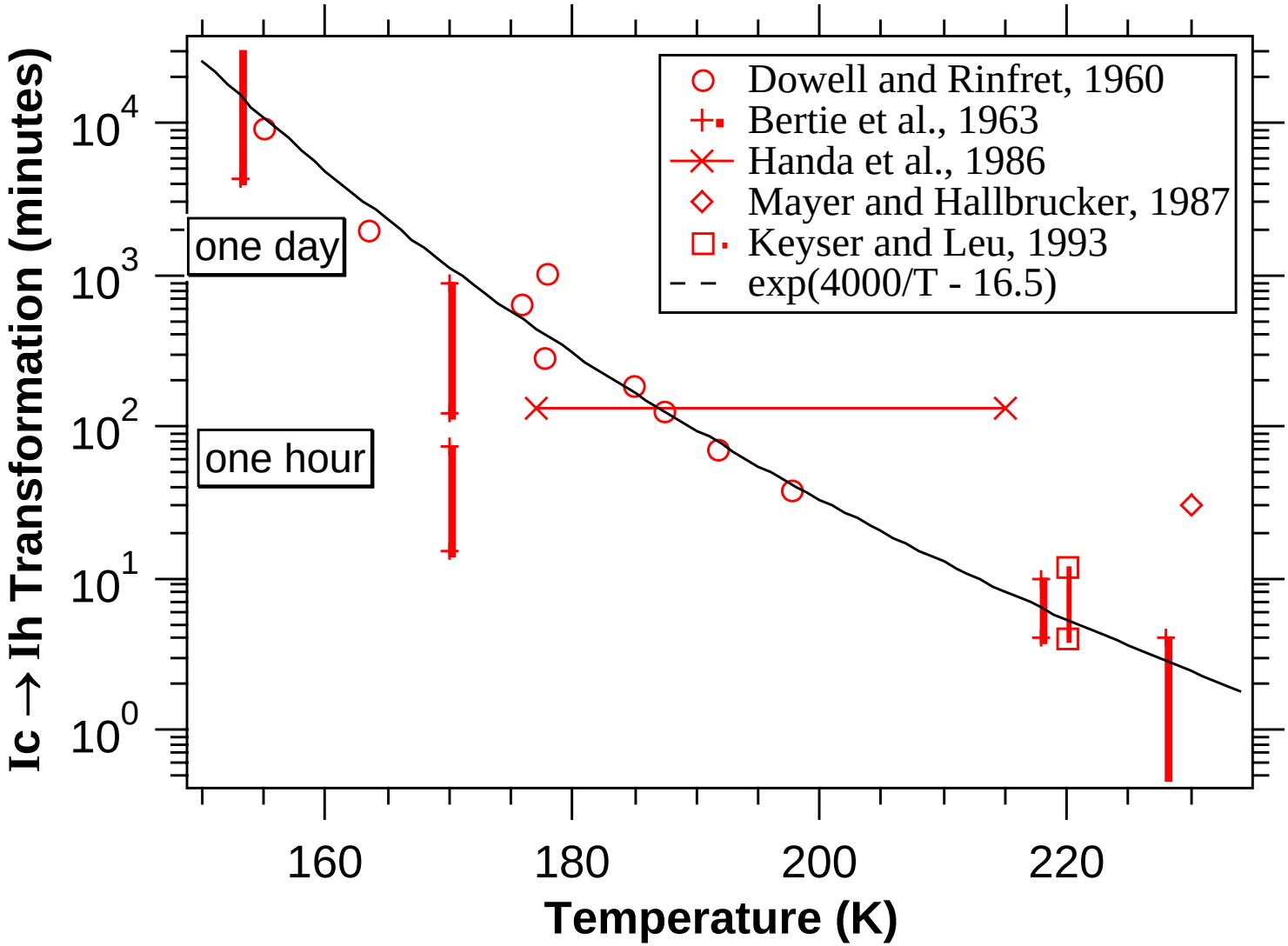


Keyser and Leu
Vapor deposited water on glass

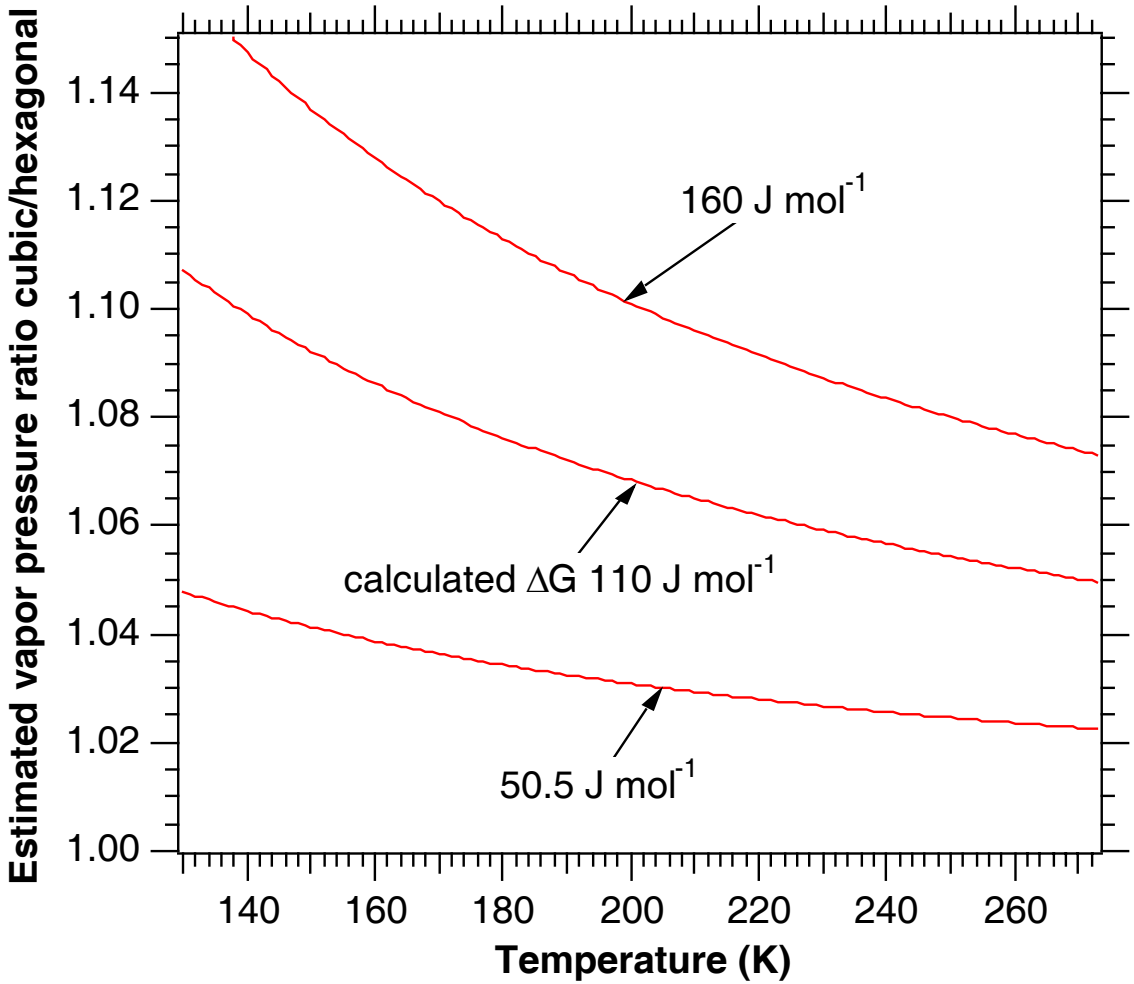
26 minutes later



Ic transforms to Ih



Relative vapor pressure of cubic ice



ΔH = 40 to 160 J mol^{-1}

(*order 1% of ice <-> liquid water*)

ΔS probably small

3 to 11% higher vapor pressure at 200 K

Why so uncertain?

- Impure samples of Ic
- Small difference

Cirrus parcel model

- Tracks freezing of aerosols, growth and transformation of Ic and Ih.
- Newly written for this problem. It gets similar answer as literature models for a published test case (Lin et al., 2002).
- 20 hour sine wave cloud plus small scale T fluctuations

Short answer:

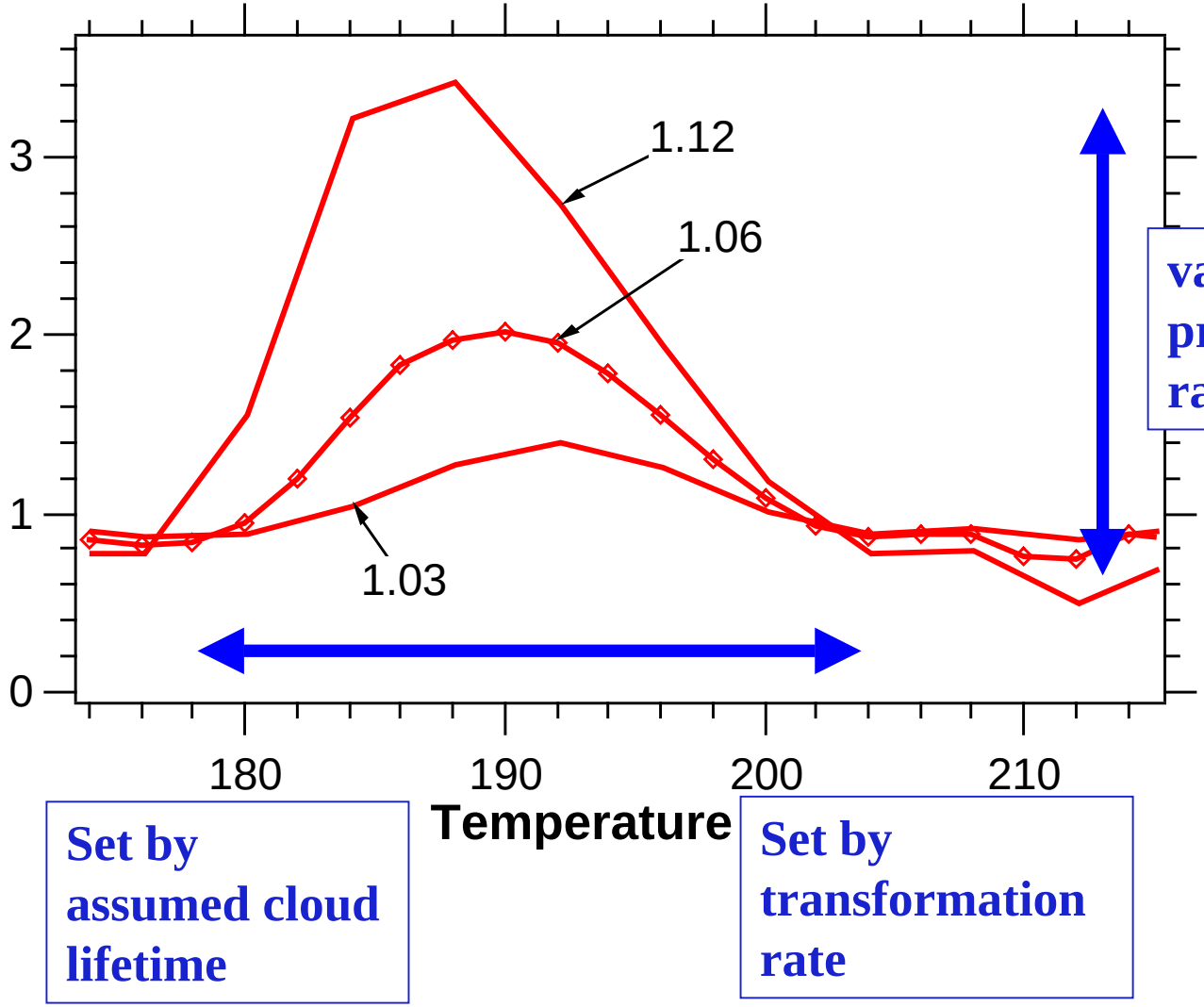
If one starts with Ic, then the first few crystals that transform to Ih grab more than their share of water.

It's the Bergeron process in slow motion.

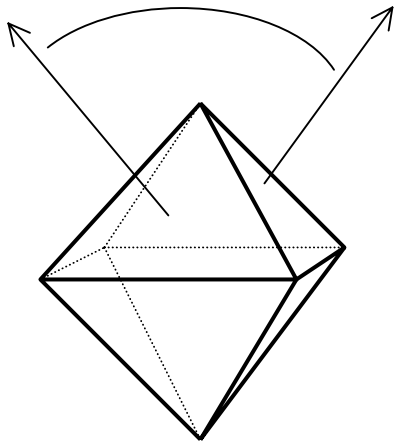
Ic => Larger particles that fall faster

Ratio of downward mass flux:

with/without cubic ice

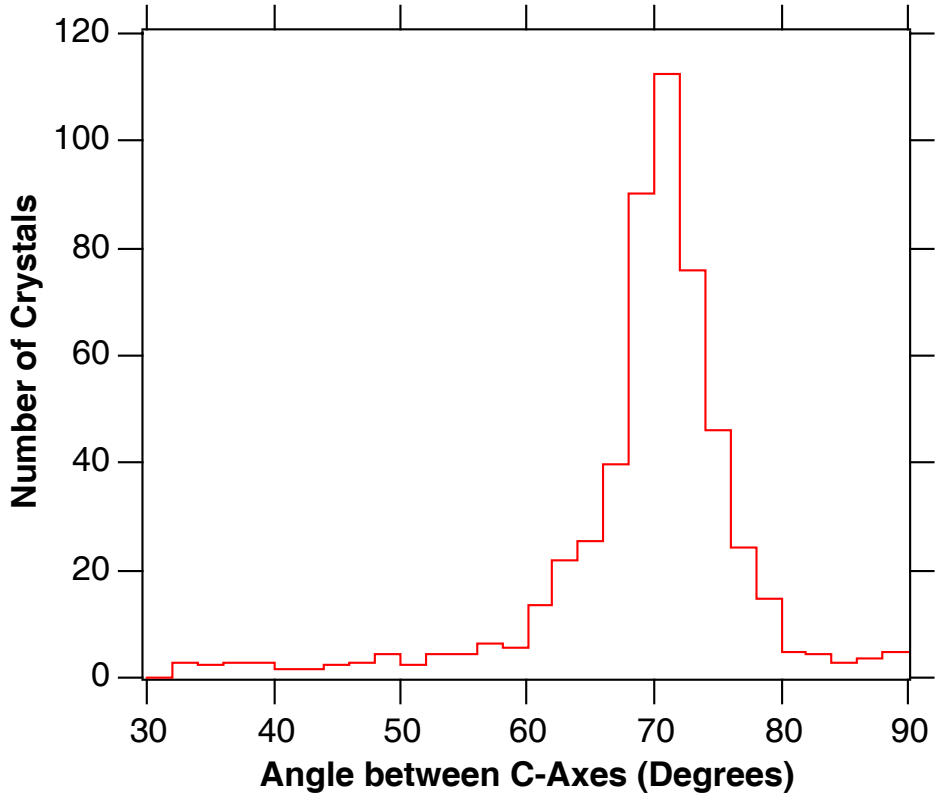


Warmer Temperatures?



Angle between faces of Ic octahedron is 70.5°

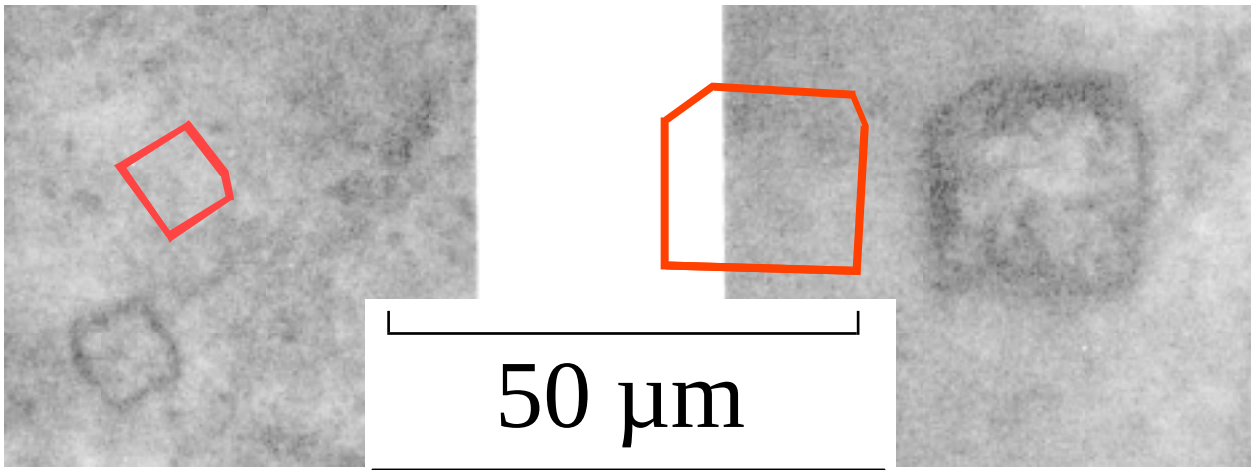
Furukawa, 1982, "spatial dendrites" formed at about -15 C



Possible explanation: hexagonal ice grew on transient cubic nucleus

Predictions (below 200 K)

1. Cubic ice crystals present for several hours.



~25% crystals in Antarctic PSCs were cubes (Goodman et al., 1989)

2. Water vapor in clouds < 200 K sometimes supersaturated with respect to Ih by 3 to 11%

Conclusions

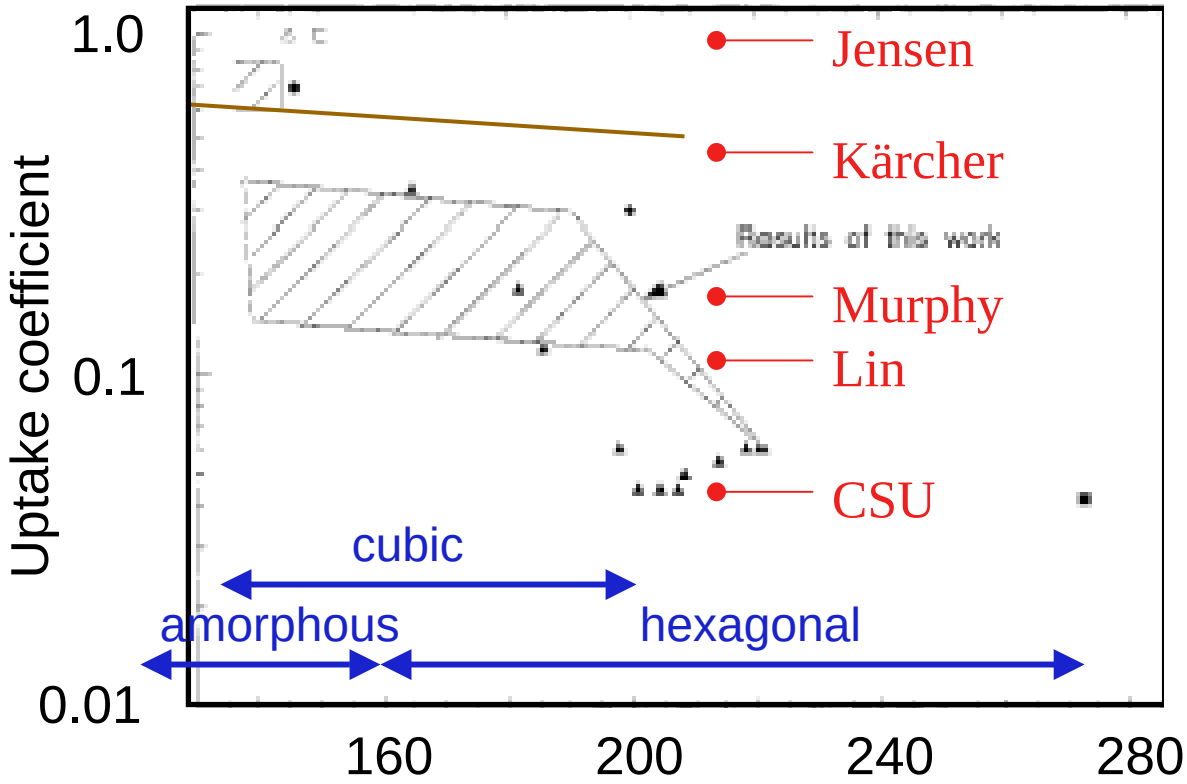
1. Laboratory studies indicate that water can freeze first to cubic ice, especially below 200 K.

2. Cubic ice can lead to larger particles that fall faster. *This happens any time a metastable phase coexists with a stable phase.*

   > ice & liquid water 0 to -40°C
   > NAT & supercooled ternary solutions in PSCs
   > Ic & Ih in cold cirrus

3. The effect on clouds is largest at about 188 K: tropical tropopause.

Uptake coefficient on ice



Factor of > 10
at ~200 K!

Figure 11. Comparison of $D_2{}^{18}O$ uptake coefficients measured in this work (hatched area) with literature values: (■) Tolbert and Middlebrook (1990);[37] (△) Brown et al. (1996);[12] (●) Koros et al. (1962);[10] (▲) Isono (1968);[36] (◆) Leu (1988);[14] (■) Delaney et al. (1964);[42] (▼) Sinarwalla et al. (1975).[43]

—— *Haynes et al., 1992*

Chaix et al., 1998