Atmospheric Ice Nucleation
Ice clouds and mixed-phase clouds

- Most precipitation that reaches the ground originates from the ice phase
- In the Tropics, precipitation via the ice phase accounts for \( \sim 70\% \) of the total precipitation amount (Lau & Wu, 2003)
- Ice can exist in clouds at temperatures close to 0°C, while liquid can exist at temperatures as cold as -40°C
- Mixed-phase clouds (i.e., co-existence of ice and liquid) can occur between -40°C and 0°C
- Information about cloud thermodynamic phase mainly comes from in situ aircraft studies, but large variability exist between different data sets
Mixed-phase clouds

Cloud phase reported from field campaigns:

Figure 8.1: Comparison of the fraction of ice, liquid and mixed-phase clouds from different studies. Note that the left-hand and right-hand y-axes are in opposite order. The gray line labelled 1 and the black solid line labelled 2 refer to the left-hand axis. All other curves refer to the right-hand axis (Korolev et al., 2003). The fraction of mixed-phase clouds can be inferred as the difference between the two gray lines and the two thick solid black lines.
Mixed-phase clouds

Cloud phase observed by Spaceborne lidar (Storelvmo and Tan, 2015):

![Graph showing supercooled cloud fraction (SCF) vs. temperature for different regions: Dust Belt, Southern Ocean, and Global Average. The graph includes error bars for each data point.]
Ice initiation in clouds

- Once a cloud extends to altitudes where $T < 0^\circ C$, ice crystal may form either by
  a) Freezing of cloud droplets (liquid $\rightarrow$ ice)
  b) Deposition of vapor to the solid phase (vapor $\rightarrow$ ice)
- In principle, in both cases both homogeneous nucleation (i.e. without the aid of an ice nucleus (IN)) and heterogeneous nucleation (with the help of an IN) are possible
- However, theory predicts that homogeneous deposition can only occur for extreme conditions of supersaturation which never occur in the atmosphere.
Homogeneous nucleation

- Homogeneous freezing in a pure liquid drop occurs when fluctuations in the molecular arrangement of the liquid produce a stable, ice-like structure that can serve as an IN.
- The probability of occurrence for such an embryonic ice crystal depends on temperature and droplet size.
- Analogous to cloud droplet formation, the embryonic ice crystal will grow spontaneously once it reaches a certain critical size.
Homogeneous nucleation II

- Below 0°C, the Gibbs free energy is lower for ice than for liquid, but formation of the new interface between liquid and ice increases the Gibbs free energy

\[
\Delta G_{i,w} = -\frac{4\pi r^3 R_v T}{3\alpha_i} \ln \frac{e_s}{e_i} + 4\pi r^2 \sigma_{i,w}
\]  

(1)

- The nucleation rate \( J \) describes the number of nucleation events per unit time and unit volume of the parent phase

\[
J = K \exp\left(-\frac{\Delta G}{kT}\right)
\]

(2)

where \( \Delta G \) is the Gibbs free energy for the phase change, \( k \) is the Boltzmann constant, and \( K \) is a kinetic prefactor.

- At -38°C, \( J \) is large enough for at least one nucleation event to be likely for typical droplet sizes within a cloud lifetime.
Heterogeneous nucleation

Four modes of nucleation are recognized for ice nuclei:

Only a small subset of atmospheric aerosol particles have the ability to act as IN. Aerosol properties that favor ice nucleation:

- Insolubility
- Size (the larger the better)
- Crystal structure similar to water
- Surface irregularities ("active sites")
Heterogeneous nucleation II

- All ice formation at temperatures warmer must occur via heterogeneous nucleation
- Laboratory measurements of ice nucleation have shown that primary biological particles, dust and soot have the ability to act as IN in the atmosphere
Heterogeneous nucleation III

- In heterogeneous ice nucleation, the IN lowers the energy barrier for the formation of a critical ice germ
- This can be visualized in terms of a contact angle, under the assumption that the ice embryo is sitting as a spherical cap on the surface of the IN
- The smaller the contact angle, the better the IN

![Diagram of contact angle between ice embryo and IN surface]
Heterogeneous nucleation IV

Reduction in Gibbs free energy barrier for IN with $\alpha=45^\circ$ and $\alpha=90^\circ$

![Graph showing the reduction in Gibbs free energy barrier for different conditions of heterogeneous nucleation.](image)

- Blue line: homogeneous nucleation
- Red line: heterogeneous nucleation, good IN (contact angle of 45°)
- Pink line: heterogeneous nucleation, moderate IN (contact angle of 90°)

The dashed vertical line indicates the critical radius $r_{\text{crit}}$ (Lüönd, 2009).
The Wegener-Bergeron-Findeisen process

- Because of the difference in saturation vapor pressure over water vs. ice, an ice crystal forming in a liquid cloud will grow rapidly at the expense of the surrounding cloud droplets.

- The environment will remain favorable for ice crystal growth by diffusion and deposition as long as cloud droplets are available to evaporate.
Diffusional growth I

- Growth of ice crystals by diffusion is analogous to that of cloud droplets, but with a complication due to non-sphericity:

\[
\frac{dm}{dt} = \frac{4\pi C(S_i - 1)}{(L_s R_v T - 1) \frac{L_s}{KT} + \frac{R_v T}{e_i(T)D_v}}
\]

- \(S_i\) is the saturation ratio with respect to ice and \(L_s\) is the latent heat of sublimation.

- Analogous to droplet growth, the two terms in the denominator represent the thermodynamic term and vapor diffusion term, respectively.

- \(m\) is the mass of the ice crystal, \(D_v\) is the molecular diffusion coefficient and \(C\) is a parameter dependent on the ice crystal size and shape (for spheres, \(C = r\))
Diffusional growth II

- Ice crystals forming in a supercooled cloud are growing by diffusion under high supersaturations, and can reach many tens of micrometers in a few minutes. In contrast to cloud droplets, they can reach the surface as individual ice crystals.

- But vapor molecules cannot unite with an ice crystal in any random way, but must join up, molecule by molecule, in such a manner that the crystal pattern is maintained.

- Because of this, the growth rate of an ice crystal will be smaller than predicted by Equation 1.

- To take this effect into account, an accommodation coefficient \(0 < \alpha_m < 1\) is introduced

\[
\frac{dm_i}{dt} = \alpha_m \frac{dm}{dt}
\]
Ice in the atmosphere
Homogeneous nucleation
Heterogeneous nucleation
Diffusional growth
Growth by accretion
Summary

Ice crystal shapes

Temperature and supersaturation controls not only the growth rate of the ice crystals, but also the ice crystal shape, or *habit*:
Growth by accretion I

- Accretion: A large precipitation particle overtakes and captures a smaller one
- When cloud droplets are captured by a falling snow crystal, rimed crystals or graupel are formed if the droplets freeze immediately upon contact
- If freezing is not immediate, denser structures form (e.g. hail)
- Aggregation is the clumping together of ice crystals to form snow flakes.
Growth by accretion II

- The equation for growth by accretion in which the collector particle is an ice particle is analogous to that of a rain drop:

\[
\frac{dM}{dt} = \bar{E} M_l \pi R^2 (v_T(R) - v_T(r))
\]  

(5)

where \( m_i \) is the mass of the ice crystal, and \( M_l \) is the liquid water content (graupel/hail formation) or ice water content (aggregate formation)

- \( v_T(R) \) is the collector fall speed, \( v_T(r) \) the fall speed of the particle being collected and \( \bar{E} \) the mean collection efficiency.

- Empirical fall speeds are of the form

\[
v_T = a D_i^b
\]

(6)

where \( a \) and \( b \) are shape dependent constants and \( D_i \) is the melted diameter.
Summary of growth processes

(a) Condensation
(b) Deposition
(c) Collision-coalescence
(d) Aggregation
(e) Rimming
(f) Bergeron-Findeisen process