Chapter 6
Cloud Microphysics
Nucleation of water vapor condensation

Clouds form when air becomes **supersaturated** with respect to liquid water/ice

Typically occurs with **ascent of air parcel** – expansion - cooling

Water vapor condenses onto some of the **particles** present in the air

Form a cloud of small water droplets or ice particles

From Aerosol 0.01 μm -> cloud particle 10μm-> raindrop 1000 μm
All in about 10 minutes!!
Cumulus Life Cycle

• Cumulus stage
  – Still fairly shallow
  – All updraft, all liquid water
  – May reach temperatures colder than 0°C

• Mature stage
  – Near maximum height
  – Mixed ice and water
  – Rain reaches ground
  – Some downdrafts

• Dissipating stage
  – Collapsing
  – Predominately ice where colder than 0°C
  – Predominately downdrafts
  – Raining hard

• Downdraft air forms cold outflows or gust fronts that may trigger new cells
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What Happens in a Cloud?

- Updraft leads to cooling and condensation.
- Forms cloud droplets, 10-50 μm in diameter.
- Cloud droplets collide and coalesce to form rain.
- Are also carried upward to temperatures < 0°C.
- Many eventually freeze.
- Ice grows faster than supercooled liquid H₂O.
- Forms snow, graupel, and hail.
- Which may melt to form rain if it falls below 0°C level.
- Precipitation loading and evaporative cooling force downdrafts.
Graupel and Hail
Nucleation

• Usually refers to the initial formation of a droplet

• More general definition: AMS Glossary

• **Homogeneous nucleation**
  – Hypothetical, droplet spontaneously forms in pure air
  – No particles present

• **Heterogeneous nucleation**
  – Droplets form on particles called cloud condensation nuclei (CCN)
Theory of Homogeneous Nucleation of Condensation

• Hypothetical, pure water without particles
• Chance collision of water vapor molecules from a embryonic water droplet at constant T and P
  – Droplet has volume (V), surface area (A)
  – Formation of a curved water surface requires energy
  – Vapor and Water molecules have Gibb’s free energies μ_l and μ_v
• Nucleation depends upon the Gibbs Function, or Gibbs Free Energy:
  – Phase with the lowest G is the one that will exist
• In thermodynamic equilibrium, ΔG = 0
\[ \Delta G = \Delta H - T \Delta S \]

- Gibb’s free energy (Available energy) can be used to examine the thermodynamic stability of the state of a system \( \Delta G = \Delta H - T \Delta S \),
- Idea, need favorable conditions for spontaneous reactions \( \Delta G < 0 = \text{spontaneous} \)
- \( \Delta H < 0 \) *exothermic*, flow of heat out of system
- \( \Delta S > 0 \) increases disorder
- \( \Delta H(\text{-}) \) energetically favorable
- \( \Delta S(\text{+}) \) entropically favorable
- \( \Delta G = \Delta H - T \Delta S, \Delta G = (-)-(+)=(-)\),
- \( \Delta G < 0, \textbf{Exergonic} - \text{Favorable for SPONTANEOUS reactions} \)
\[ \Delta G^\circ = \Delta H^\circ - T \Delta S^\circ \]

• Gibb’s free energy (Available energy) can be used to examine the thermodynamic stability of the state of a system \( \Delta G = \Delta H - T \Delta S \),
• Idea, need favorable conditions for spontaneous reactions
• \( \Delta H > 0 \) endothermic, absorbs heat
• \( \Delta S < 0 \) decreases entropy
• \( \Delta H(+) \) energetically unfavorable
• \( \Delta S(-) \) entropically unfavorable
• less favorable for spontaneous reaction
  • \( \Delta G = \Delta H - T \Delta S \), \( \Delta G = (+) - (-) = (+) \)
  • \( \Delta G > 0 \), Endergonic, NON-SPONTANEOUS
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Theory of Homogeneous Nucleation of Condensation

The energy required (or released) to condense a drop is the difference in work done to create a unit area of vapor-liquid interface ($A\sigma$) and the difference between Gibbs energy ($\mu$ in book) between the vapor and liquid.

$$\Delta E = A\sigma - nV(\mu_v - \mu_l)$$

- Net increase in energy of the system due to droplet formation
- Work to create surface area of droplet
- Decrease in Gibbs free energy due to condensation

$n$=# of molecules/unit volume of liquid
$A\sigma$= Area * work required to create a unit area of vapor liquid interface ("interfacial energy" or "surface energy")
$V$=Volume
The Phase That Exists Will Have the Minimum Gibbs Function

For water vapor:
\[ G_V = kT \ln \frac{e}{e_s} = \mu_v - \mu_l \]
Per molecule

\( e \), vapor pressure
\( T \), temperature of the system
\( e_s \), is the saturation vapor pressure over a plane surface at \( T \)
\( k \), Boltzmann constant (JK\(^{-1}\))
Theory of Homogeneous Nucleation of Condensation

\[ \Delta E = A \sigma - nVkT \ln \frac{e}{e_s} \]

- \( \Delta E \) is the net increase in the energy of the system due to water droplet formation
- \( \Delta E \) Function of radius \( R \), \( T \), and \( RH \)

\[ \Delta E = 4\pi R^2 \sigma - \frac{4}{3}\pi R^3 n kT \ln \frac{e}{e_s} \]

- Under subsaturated conditions (ie vapor pressure \( e \) < \( e_s \)), \( \ln(e/e_s) \) is negative and \( \Delta E \) is always positive and increases with increasing droplet radius
- Larger the drop, the greater the increase in \( \Delta E \) of the system
- Recall, A system reaches equilibrium by reducing its energy
- Droplet formation is not favored in subsaturated conditions

\( k = \)Boltzmann constant \((JK^{-1})\)
Theory of Homogeneous Nucleation of Condensation

\[ \Delta E = A\sigma - n\sqrt{\frac{e}{e_s}} \]

- \( \Delta E \) is the net increase in the energy of the system due to water droplet formation
- \( \Delta E \) Function of radius \( R \)

\[ \Delta E = 4\pi R^2\sigma - \frac{4}{3}\pi R^3 nkT \ln \frac{e}{e_s} \]

- Under supersaturated conditions (ie vapor pressure \( e > e_s \)), \( \ln(e/es) \) is positive, and \( \Delta E \) can be positive or negative depending on the Radius (\( R \))

\( K = \text{Boltzmann's constant} = R^* / N_A \)
\[ \Delta E = 4 \pi R^2 \sigma - \frac{4}{3} \pi R^3 n k T \ln \frac{e}{e_s} \]

- Under subsaturated conditions (ie vapor pressure \( e < e_s \)), \( \ln(e/e_s) \) is negative and \( \Delta E \) is always positive and increases with increasing droplet radius.
- Larger the drop, the greater the increase in \( \Delta E \) of the system.
- Under supersaturated conditions (ie vapor pressure \( e > e_s \)), \( \ln(e/e_s) \) is positive, and \( \Delta E \) can be positive or negative depending on the Radius.

\[ r = \frac{2 \sigma}{n k T \ln \frac{e}{e_s}} \]

Increase \( \Delta E \) in the energy of a system due to the formation of a water droplet of radius \( R \) from water vapor with pressure \( e \); \( e_s \) is the saturation vapor pressure with respect to a plane surface of water at the temperature of the system.
Hence, under supersaturated conditions, embryonic droplets with $R < r$ tend to evaporate, since by so doing they decrease $\Delta E$.

However, droplets that manage to grow by chance collisions to a radius that just exceeds $r$ will continue to grow spontaneously by condensation from the vapor phase, since this will produce a decrease in $\Delta E$.

At $R=r$, a droplet can grow or evaporate infinitesimally without any change in $E$.

Find $r$ by setting $d(\Delta E)/dR=0$ at $R=r$

$$r = \frac{2\sigma}{nkT \ln \frac{e}{e_s}}$$

**Kelvin’s equation** used to find $r$ in unstable equilibrium, given $e$

Also, can find $e_s$ for a given $r$

Increase $\Delta E$ in the energy of a system due to the formation of a water droplet of radius $R$ from water vapor with pressure $e$; $e_s$ is the saturation vapor pressure with respect to a plane surface of water at the temperature of the system.
pure water droplet of radius 0.01\(\mu\text{m}\) requires a relative humidity of 112\% (i.e., a supersaturation of 12\%) to be in (unstable) equilibrium with its environment,

while a droplet of radius 1\(\mu\text{m}\) requires a relative humidity of only 100.12\% (i.e., a supersaturation of 0.12\%).
Heterogeneous nucleation

- Natural super-saturations are too low for growth of droplets with $r < 0.1 \mu m$,
- **Heterogeneous nucleation** takes place on wettable particles
- Heterogeneous nucleation require lower supersaturations
- Particle (wettable) at 0.3$\mu m$, a film of water forms and this film will be in **unstable equilibrium** at 0.4% supersaturation
- Size will increase at supersaturation levels $> 0.4\%$
Kinds of Condensation Nuclei

• For **hydrophyllic** (wettable) nuclei, water attaches to the surface.
• For **hydrophobic** nuclei, water may bead on the surface,
  – But doesn’t attach
  – Not good condensation nuclei
• For **hygroscopic** nuclei, water soaks into the surface and eventually dissolves the nucleus.
Kinds of Condensation Nuclei

• CCN concentration
  – Over land, 100-1000 CCN cm\(^{-3}\) typically become active at 1% supersaturation
  – Over the oceans, <100 CCN cm\(^{-3}\) typically become active at 1% supersaturation
  – Over the Arctic, <10-50 CCN cm\(^{-3}\) typically become active at 1% supersaturation

The larger the (wettable) particle, more readily wetted
Increase the solubility, reduce the supersaturation required for it to become a CCN
Heterogeneous Nucleation

- **Hygroscopic** CCN are particularly effective condensation initiators
  - Generally made of soluble salts
- When droplet forms, solution has a much lower vapor pressure than pure water
  - Condensation begins when RH < 100%
- Droplet growth requires supersaturations of less than 1%
  - Such supersaturations are achieved in updrafts
  - Köhler Curves give the equilibrium droplet size for a given RH.
Suppose RH = 100.1%

Numbers indicate mass of dissolved salt (NaCl)

"Saturation ratio" = RH/100

Köhler Curves
Droplets grow until they reach equilibrium radius.
Droplets grow until they reach equilibrium radius.
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Droplets grow until they reach equilibrium radius.
Droplet Growth

• If ambient RH < value at peak of curve, droplets stop growing when much smaller than typical cloud drop

• They are called *haze* droplets
Suppose RH = 100.3%
Droplets growing on smaller nuclei behave as before.
Look at largest nucleus

Saturation Ratio

Radius (μm)

$10^{-19}$ g

$10^{-18}$ g

$10^{-17}$ g
Droplet keeps growing!
Droplet “Activation”

• If ambient RH > peak value, droplet grows indefinitely
• Once droplet has gotten “over the hump”, it is said to be activated.
• Rate of droplet growth decreases as droplets grow
• Let \( r \) = droplet radius
• It can be shown that \( \frac{dr}{dt} \propto \frac{1}{r} \)
Heterogeneous nucleation

• Some particles dissolve and create a solution droplet (not pure water droplet)
• $e_s$ for solution droplet is less than for pure water
• Fractional reduction in water vapor pressure given by Raoult’s law $e'/e_s = f$
• Where $e'$ is saturation vapor pressure next to solution droplet with molar fraction $f$ of pure water (i.e., moles pure/total moles)
• So that the required supersaturation is reduced for solutions
Heterogeneous nucleation

• So that the required supersaturation is reduced for solutions
• As droplet grows solution dilutes
• **Kholer Curves** reflect both lower interfacial energy and dilution of the solute as the droplets grow.
• As $r$ increases, more pure water and dilution, RH next to droplet same as that next to pure water droplet
• At A $(\text{NH}_4)_2\text{SO}_4$ reaches unstable equilibrium, NaCl will continue to grow

Unactivated, haze droplet
Activated, fog or cloud

Curve 2 $10^{-19}$ kg of NaCl
Curve 5 $10^{-19}$ kg of $(\text{NH}_4)_2\text{SO}_4$
Heterogeneous nucleation

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![Graph showing Equilibrium RH vs radius for Pure water and Salt solutions](image-url)
Clouds Begin With Atmospheric Aerosols

- Suspensions of solid or liquid in a gas, in this case air.
  - Clouds are aerosols, too, but in meteorology we usually mean different chemical composition than H2O.
  - What we’re interested in is Cloud Condensation Nuclei (CCN)
- **Sizes:** measured in μm (= 10⁻³ mm) 0.01 to 0.1 μm.
- Because of drafts and low mass aerosol particles fall 0.03 to 3 mm s⁻¹ (i.e. a few meters a day)
- Number activated depends upon supersaturation
- **Concentration:**
  - Oceans 50-100 cm⁻³
  - Inland (normal) 300-1000 cm⁻³
  - Inland (drought) >1000 cm⁻³
  - Concentration decreases dramatically above 2-4 km altitude
- **Sources:** combustion, sea salt, some soils, decomposition of some plants
- **Sinks:** Coagulation, capture in clouds, fallout, precipitation scavenging

At 1% supersaturation the radius of insoluble vs soluble is significant!

- *Wettable nuclei > 0.1 μm*
- *Soluble nuclei > 0.01 μm*
- Most CCN are *mixed nuclei*, partially soluble, partly insoluble
Observed CCN Concentrations

Land has higher concentration (with smaller radius) of CCN than Marine
ABL has 5 x more CCN than FA over land
ABL has about the same as FA over marine regions
Land surface is likely source for CCN.
Soil and dust (small)
  Wildfires (large)
diesel engines (large)
Sea Salt over oceans, though not dominant

Warm Clouds
Microphysical Parameters

• Liquid water content (LWC)
  – grams of liquid water per m$^3$ of cloud

• Droplet concentration, N
  – Number of droplets per cm$^3$

• Mean droplet size, $\bar{r}$
  – Usually given in µm

• Not independent – knowledge of any two determines the third
Warm Cloud Structure

Occur below the 0°C isotherm in regions of updrafts.

High LCW follow strong updrafts, note downdrafts at boundaries.

Inhomogeneous, regions of high LWC interspersed with regions of very little LWC.

Droplet spectrum ranges from ~5 μm to 25 μm.

Measured from an aircraft as it flew in a horizontal track across the width and about half-way between the cloud base and cloud top in a small, warm, nonraining cumulus cloud. The cloud was about 2km deep.
Warm Cloud Structure

Marine (cumulus) clouds
Lower concentrations above 100 cm\(^{-1}\)
None with concentration larger than 200 cm\(^{-1}\)

Continental (cumulus) clouds (higher CCN)
Most concentrations ~ 300 cm\(^{-1}\)
Some concentrations > 900 cm\(^{-1}\)
MORE CCN over land!

LCW nearly the same for land and marine so
Fewer drops Marine, same LWC per cloud
Larger cloud droplets in Marine
Smaller cloud droplets in continental
Warm Cloud Structure

Cloud optical thickness ($\tau_c$) and cloud droplet effective radius ($r_e$)

$r_e$ generally smaller over land
Ship tracks in clouds
CLW content and Entrainment

Once air is lifted above Lifting Condensation Level, we can determine the amount of liquid water using Skew T-InP chart, *adiabatic liquid water content*

Measurements of LWC, however, are well below adiabatic LWC

Due to **entrainment of unsaturated air**

*LWC evaporates to saturate the entrained air*

Air entrained at the top of cloud is distributed down

Evaporation cools parcel, if parcel isn’t mixed (ie not a parcel any more) it sinks and interacts with cloud at lower level.

Parcels may descend kilometers (Penetrative downdrafts)

Thus, the patchy distribution of LWC

---

LWC measured from an aircraft as it flew in a horizontal track across the width of a small, warm, cumulus cloud.
Marine Sc entrainment

**LW Cool** to space at top and **LW heated** from below causes convection
Cool cloudy air sinks (evaporates)
Warm cloudy air rises and droplets grow

- Entrainment of FA air into ABL
- Cooling by evaporation of cloud water drives parcel downward

Thus the cellular appearance of stratocumulus clouds
Processes for Cloud Droplet Growth

• How does this happen??
• By:
  – condensation
  – collision/coalescence
  – ice-crystal process
Water Droplet Growth - Condensation

Flux (diffusion) of vapor to droplet *(schematic shows “net flux” of vapor towards droplet, i.e., droplet grows)*

Need to consider:

Vapor flux due to gradient between saturation vapor pressure at droplet surface and environment (at $\infty$). 

$$\frac{dM}{dt} = 4\pi x^2 D \frac{d\rho_v}{dx}$$

Where $D$ *diffusion coefficient* of water vapor is rate of mass flow of wv across (and normal to) a unit area in the presence of a unit gradient in water vapor density $\rho$

Effect of Latent heat effecting droplet saturation vapor pressure (equilibrium temperature accounting for heat flux away from droplet).
Water Droplet Growth - Condensation

Recall, condensation decreases as radius increases

\[
\frac{dr}{dt} \propto \frac{1}{r}
\]

Updraft, supersaturation, activation of CCN,
When rate of wv (supersaturated) = rate of condensation of wv on to CCN, max cloud droplet concentration is reached

- Further droplet growth reduces supersaturation
- Smaller haze droplets evaporate
- And size of droplets become uniform (*monodispersed distribution*)

Works near the base of a cloud, **maximum size ~10μm**, but small size and decreasing rate of growth suggest **condensation insufficient to create raindrops**.

From cloud r=10μm to raindrop r=1mm a millionfold increase in volume, only need 1 in a million cloud drops to reach r=1mm for a cloud to rain.
Water Droplet Growth
Condensation & Collision

- **Condensational growth**: diffusion of vapor to droplet
- **Collisional growth**: collision and coalescence (accretion, coagulation) between droplets
- **Terminal Fall Speed**: is a function of radius, larger drops fall faster
  Note at larger r (mms), drag tends to increase, and to reduced sphericity (increase drag, reduce terminal velocity)
- As collector drop \((r_1)\) approaches smaller drop \((r_2)\), we can define an effective collision cross section \((y)\) and a collision efficiency \(E\)

\[
E = \frac{y^2}{(r_1 + r_2)^2}
\]

- Collision efficiency \((E)\) increases with increasing \(r_1\), \(E\) is small for \(r_1\) less than 20\(\mu m\).
  \(E\) is small if \(r_1\) much larger than \(r_2\), ie \(r_2\) will follow streamlines and avoid collision
- As \(r_2/r_1\) reaches 0.6-0.9 **comparable fall speeds** reduce \(E\)
- As \(r_2/r_1\) reaches 1 wake effects act to increase \(E\)
Water Droplet Growth
Condensation & Collision

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- As \( r_2 / r_1 \) reaches 1 wake effects act to increase \( E \)

Collision is not sufficient for coalescence

Water droplets can bounce off of each other (due to a cushion of air)

Coalescence efficiency \( E' \) fraction of collisions that result in coalescence
Collection efficiency \( E_c = EE' \)
Continuous collection model

- Collector drop falls through cloud of equal sized droplets
- Droplets are uniformly distributed
- Droplets will be collected at a uniform rate for given $r_1$
  \[
  \frac{dM}{dt} = \pi r_1^2 (v_1 - v_2) w_l E_c
  \]
- Where $w_l$ is the Liquid Water Content (in kgm$^{-3}$) of the cloud droplets $r_2$
- If we assume $v_1 >> v_2$ and coalescence efficiency is unity $E_c = E$, we can define
  \[
  \frac{dr_1}{dt} = \frac{v_1 w_l E}{4 \rho_l}
  \]

Because $v_1$ and $E$ increase as $r_1$ increases, it follows that $\frac{dr_1}{dt}$ increases with increasing $r_1$

The growth of a drop by collection is an acceleration process!
Water Droplet Growth - Collisions

- Droplets collide and coalesce (accrete, merge, coagulate) with other droplets.

- Collisions governed primarily by different fall velocities between small and large droplets (ignoring turbulence and other non-gravitational forcing).

- Collisions enhanced as droplets (r1) grow and differential fall velocities increase.

- Not necessarily a very efficient process (requires relatively long times for large precipitation size drops to form).

- Rain drops are those large enough to fall out and survive trip to the ground without evaporating in lower/dryer layers of the atmosphere.

Because velocity (v1) and collision efficiency (E) increase as r1 increases, it follows that \( \frac{dr_1}{dt} \) increases with increasing r1.

The growth of a drop by collection is an acceleration process!
Collision/Coalescence

• Collision/Coalescence - cloud droplet growth by collision

• this is a dominant process for precipitation formation in warm clouds (tops warmer than about -15°C)

• some cloud droplets will grow large enough and will start to fall in the cloud -->>

• since the bigger drops fall faster than the smaller drops, they will "collect" the smaller drops - the bigger drop grows

• droplet fall speed is called its **terminal velocity**

• need droplets of different sizes for this process to really work

• **Q**: what determines the droplets fall speed relative to the ground??
Droplet Fall Speeds and Droplet Growth

• Q: what determines the droplets fall speed relative to the ground??
• A: droplet size and updraft strength -->
• given a growing cu with an updraft (w) strength of 4 ms\(^{-1}\):
  • if the particle terminal velocity (Vt) is -2 ms\(^{-1}\), the particles fall speed (Vg) is: -2 ms\(^{-1}\)
  • if the particle terminal velocity is -4 ms\(^{-1}\), the particles fall speed is: 0 ms\(^{-1}\)
  • if the particle terminal velocity is -6 ms\(^{-1}\), the particles fall speed is: 2 ms\(^{-1}\)
Life cycle of a droplet

• Growth by collision
• the drop initially forms in the updraft of the cloud near cloud base
• it grows in size by collisions
• since \( V_g = w + V_t \)
  – \( V_g \) = ground relative fall speed of the drop
  – \( w \) = updraft velocity
  – \( V_t \) = drop's terminal velocity
• then the drop will begin to fall when \( V_t > w \)
Factors promoting growth by collision/coalescence

- Different drop sizes
- Thicker clouds
- Stronger updrafts

- Consider a shallow stratus deck....
Droplet Growth in a Shallow Stratus Deck

- Often, drops will evaporate from shallow stratus before reaching the ground
- or you may get drizzle if they are large enough
TABLE 7.3. Distance a Drop Falls before Evaporating, assuming Isothermal Atmosphere with $T = 280 \, \text{K}$, $S = 0.8$

<table>
<thead>
<tr>
<th>Initial radius</th>
<th>Distance fallen</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 , \mu\text{m}$</td>
<td>$2 , \mu\text{m}$</td>
</tr>
<tr>
<td>$3 , \mu\text{m}$</td>
<td>$0.17 , \text{mm}$</td>
</tr>
<tr>
<td>$10 , \mu\text{m}$</td>
<td>$2.1 , \text{cm}$</td>
</tr>
<tr>
<td>$30 , \mu\text{m}$</td>
<td>$1.69 , \text{m}$</td>
</tr>
<tr>
<td>$0.1 , \text{mm}$</td>
<td>$208 , \text{m}$</td>
</tr>
<tr>
<td>$0.15 , \text{mm}$</td>
<td>$1.05 , \text{km}$</td>
</tr>
</tbody>
</table>
Mechanisms for transition from Condensation to collection coalescence

- Giant cloud condensation nuclei (GCCN) ~3μm act as embryos for collector drop increase likelihood of precipitation.

- Effects of turbulence, fluctuating super-saturations, downdrafts, vorticity lead to enhanced growth in certain areas.

- Radiative broadening, droplet radiates heat to environment, reducing saturation vapor pressure near the droplet surface, leading to growth.

- Stochastic Collection as opposed to continuous collection model with uniform distribution of same size droplets, here collisions are random but large drops are favored for growth. Provides a mechanism for developing broad droplet size spectra from uniform droplet sizes formed by condensation. Also shows how small fraction of droplets can grow much faster than average by statistically distributed collisions.