Lecture 5.

Major processes involving aerosol and clouds

Required reading:

Chs. 20 and 23 from Heintzenberg&Charlson (2009)

Additional reading:

Ghan S.J. and S. E. Schwartz, Aerosol Properties and Processes: A Path from Field and Laboratory Measurements to Global Climate Models. Bulletin of the American Meteorological Society, Volume 88, Issue 7 (July 2007) pp. 1059-1083.

Cantrell, W., and A. Heymsfield, Production of ice in tropospheric clouds: A review. BAMS,795-807, 2005.

Tao, W.-K., J.-P. Chen, Z. Li, C. Wang, and C. Zhang (2012), Impact of aerosols on convective clouds and precipitation, Rev. Geophys., 50, RG2001, doi:10.1029/2011RG000369.

Stevens, B., and G. Feingold (2009), Untangling aerosol effects on clouds and precipitation in a buffered system, Nature, 461(7264), 607-613.

Andreae, M.O., and D. Rosenfeld, Aerosol–cloud–precipitation interactions. Part 1. The nature and sources of cloud-active aerosols. Earth-Science Reviews, Volume 89, Issues 1–2, July 2008, Pages 13-41

Impacts of aerosol and clouds on climate

Direct aerosol radiative impact – perturbation of the Earth radiative balance through altering solar and terrestrial IR radiation

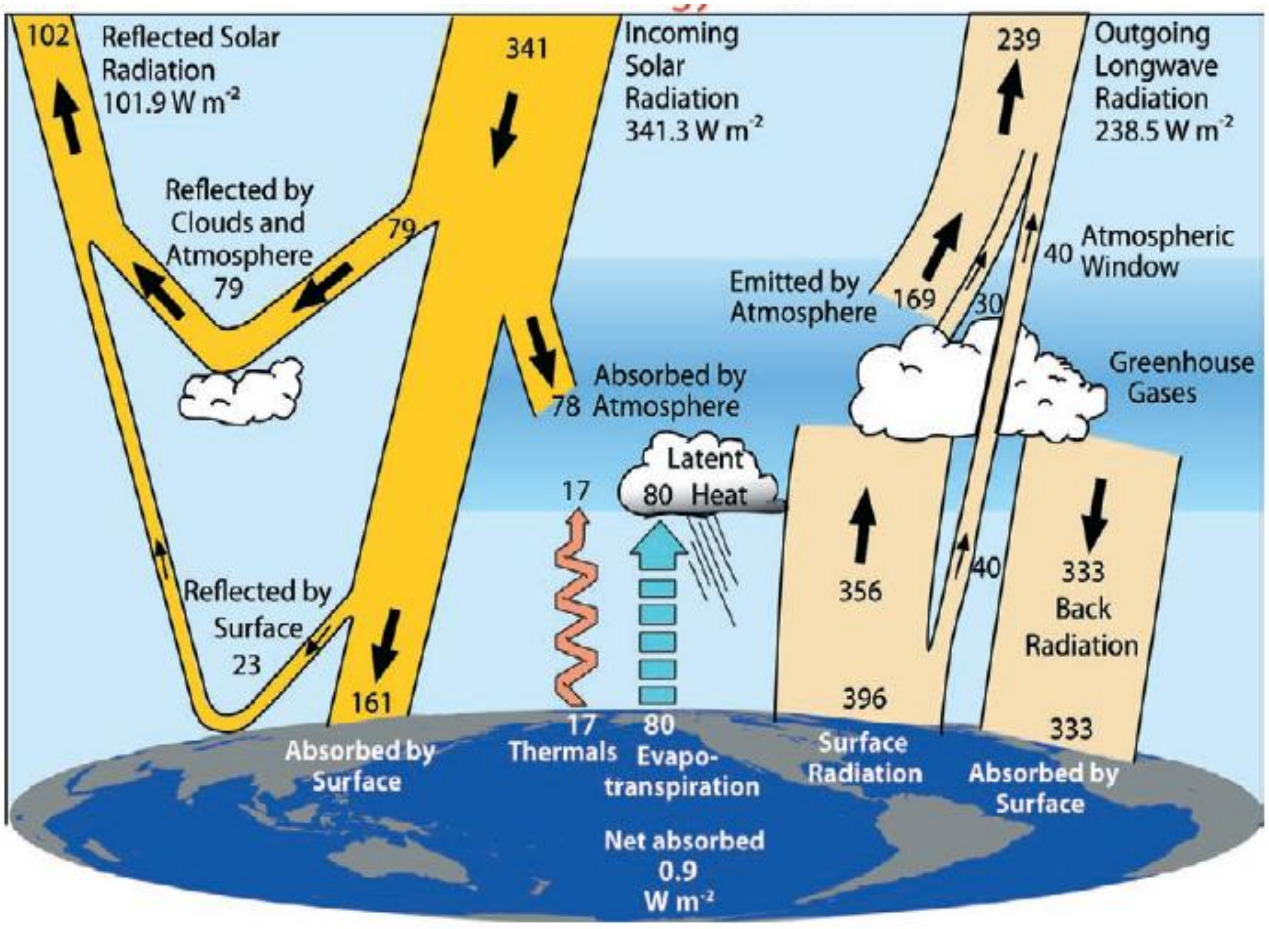


Figure 5.1 Earth's energy balance diagram from Trenberth et al. (2009). The global annual mean Earth's energy budget for the Mar 2000 to May 2004 period (W/m^2). The broad arrows indicate the schematic flow of energy in proportion to their importance.

Global mean forcing (based on GCMs)

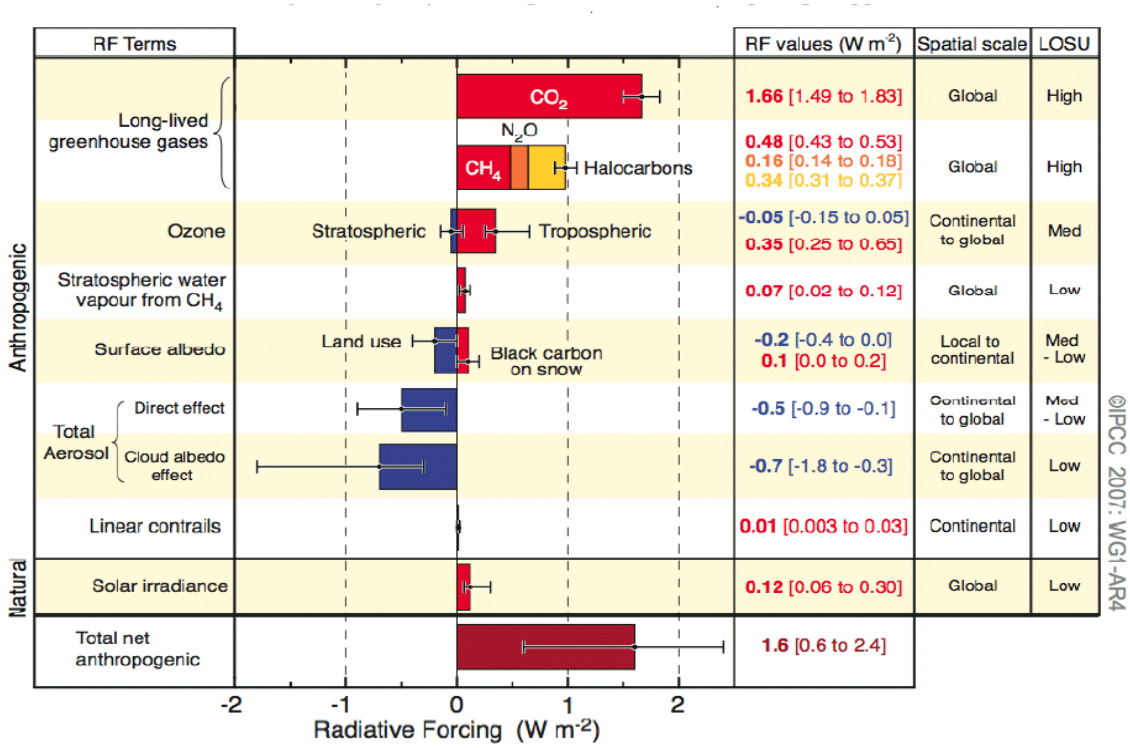


Figure 5.2 Intergovernmental Panel on Climate Change (IPCC, 2007**)**: Global mean radiative forcing (W/m2): 2005 relative to 1750

Direct radiative forcing by individual aerosol components (W/m2):

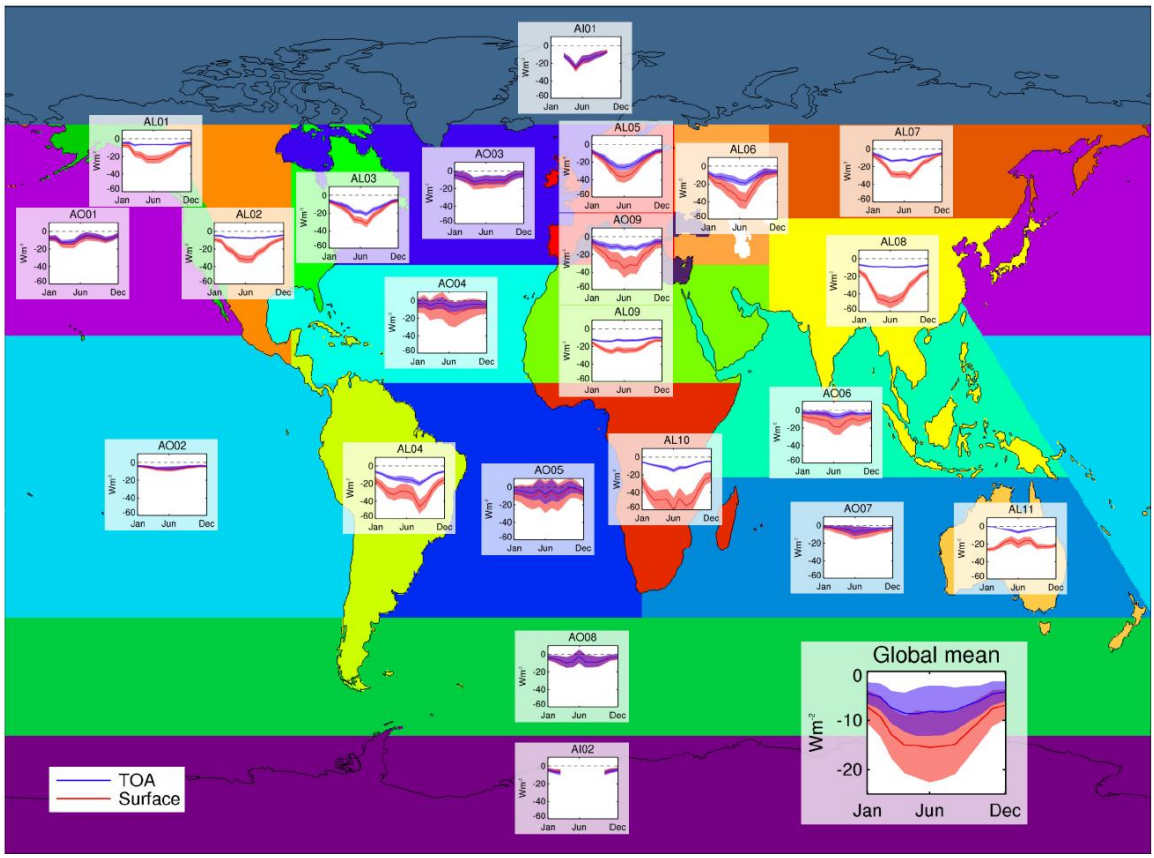
Sulfates -0.4 (+/- 0.2) Nitrates -0.1 (+/-0.2)

Fossil fuel OC -0.05 (+/- 0.005) Fossil fuel BC +0.2 (+/- 0.15)

Biomass burning +0.03 (+/-0.12) Dust -0.1 (+/-0.2) from -0.3 to +0.1

Regional distribution of aerosol radiative forcing

Example of monthly, regional estimates of the clear-sky aerosol radiative forcing (for 2006 relative to pre-industrial) based satellite aerosol optical depth retrieval and radiative transfer modeling (see Thomas et al., 2013, ACP)



Findings:

The developed industrial regions (AL01, AL03, AL05, AL06 and AO09) all show a distinct pattern in the radiative forcing where the forcing is far stronger in the summer months and approaches zero during the winter, whereas the developing world shows a much more consistent forcing throughout the year. This pattern is due to a combination of the regional variation in seasonal cycle of insolation, and the seasonal cycle in AOD. One potential contributor to the latter effect is that aerosol loading in developed regions is generally dominated by photo-chemical production of secondary aerosol, whereas the primary aerosol production, such as from biomass burning, is more important in developing regions.

Aerosol indirect effect

See Table 23.1 in Ch.23

Table 23.1 Overview of known aerosol indirect effects on net radiative flux (at TOA and at the surface) and on precipitation, and an assessment of level of current scientific understanding. Modified from Denman et al. (2007).

Effect	Cloud albedo effect	Cloud lifetime effect	Semi-direct effect	Glaciation indirect effect	Thermodynamic effect
Cloud types affected	All; greatest for clouds of intermediate optical thickness	All	All	Mixed-phase	Mixed-phase
Process	For same cloud water or ice content more but smaller cloud particles reflect more solar radiation	Smaller cloud particles decrease precipitation efficiency prolonging cloud lifetime	Absorption of solar radiation by absorbing aerosols evaporates cloud particles, increases static stability	Increase in ice nuclei increases precipitation efficiency	Smaller cloud droplets delay freezing and cause supercooled clouds to extend to colder temperatures
Change in net TOA irradiation	–	–	+/–	+	+/–
Potential magnitude	medium	medium	small	medium	medium
Scientific understanding	low	very low	very low	very low	very low
Change in surface irradiation	–	–	+/–	+	+/–
Potential magnitude	medium	medium	large	medium	medium
Scientific understanding	low	very low	very low	very low	very low
Change in precipitation	N/A	–	–	+	+/–
Potential magnitude	N/A	small	large	medium	medium
Scientific understanding	N/A	very low	very low	very low	very low

Aerosol processes:

From Ghan and Schwartz (BAMS, 2007)

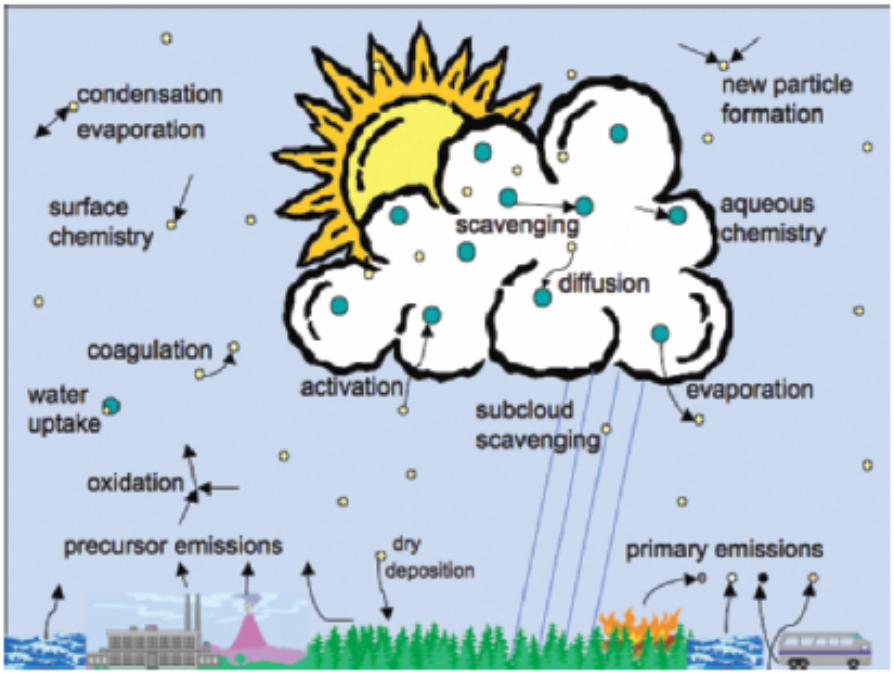
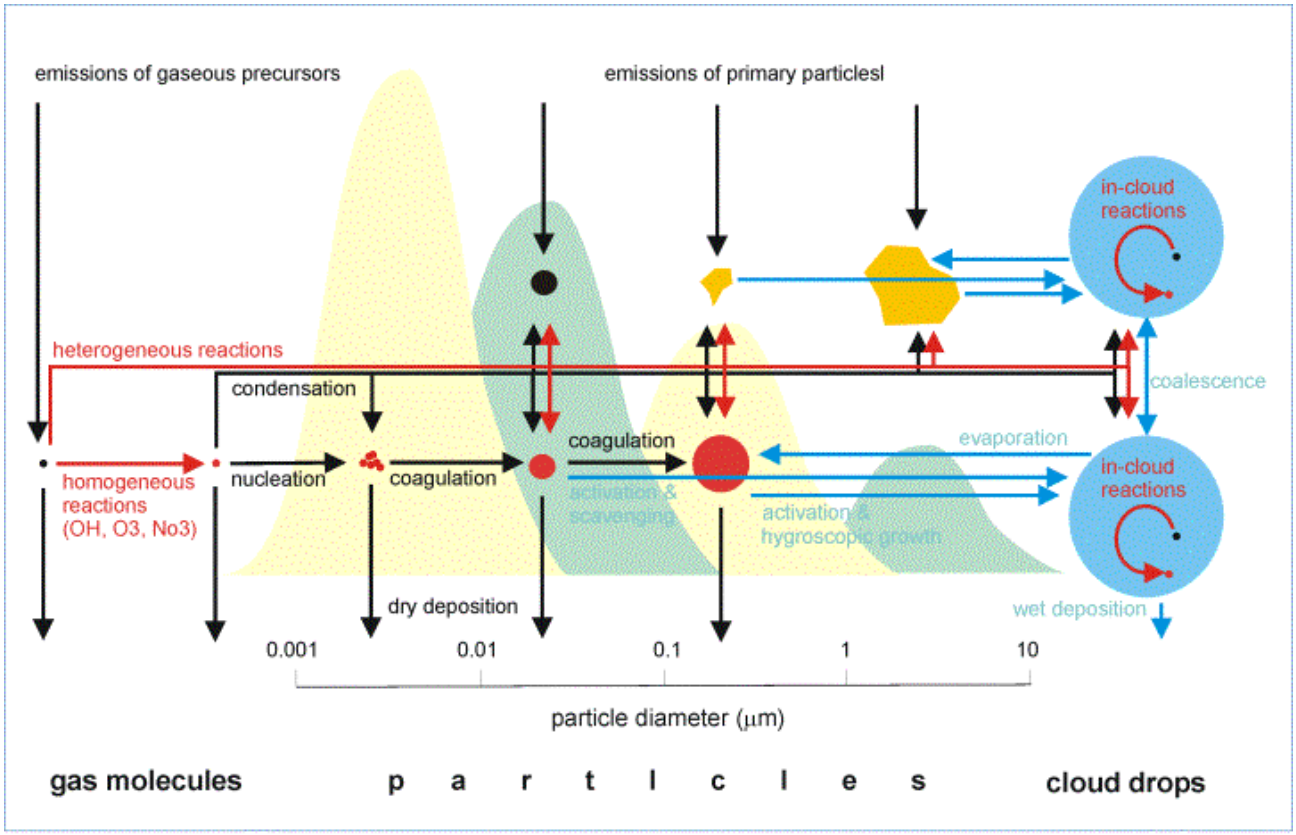


FIG. 1. Important aerosol processes that influence climate and must be accurately represented in future generations of climate models. Aerosol particles are directly emitted as primary particles and are formed secondarily by oxidation of emitted gaseous precursors. The formation of low-volatility materials in this way results in new particle formation and condensation onto existing particles. Aqueous-phase oxidation of gas-phase precursors within cloud droplets accretes additional mass onto existing particles but does not result in new particle formation. Particles age by surface chemistry and coagulation as well as by condensation. With increasing relative humidity particles may accrete water vapor by deliquescence and further hygroscopic growth; with decreasing relative humidity water is lost and ultimately particles may effloresce to the dry state. The uptake of water increases particle size, affecting also the particle optical properties. During cloud formation some fraction of aerosol particles serve as cloud condensation nuclei, by becoming activated, that is, overcoming a free-energy barrier to form cloud droplets. Within clouds interstitial particles can become attached to cloud droplets by diffusion, and activated particles are combined when cloud droplets collide and coalesce. If cloud droplets evaporate the particles are resuspended, but if the cloud precipitates the particles are carried below the cloud and reach the surface, unless the precipitating particles completely evaporate. Aerosol particles below precipitating cloud can also be removed from the atmosphere by impaction by precipitating drops and by dry deposition to the surface.



Raes et al., Atm. Env., 2000

Figure 5.3 Schematic picture of the microphysical processes that influence the number size distribution and chemical composition of the atmospheric aerosol particles (Raes, et al., 2000). A number size distribution, with nucleation, Aitken, accumulation and coarse mode, is schematized in the background, illustrating that the highest number of particles are usually found with the smallest particles.

Major aerosol processes:

removal processes:

Dry deposition

Gravitational settling

Wet removal (wash-out)

Coagulation is the sticking together of two particles. It is the result of particles coming into contact due to Brownian diffusion, turbulence or some force (electrostatic, phoretic effects *etc*.). Note that contact does not necessarily lead to coagulation, but must happen as a pre-requisite. This happens more quickly for Aitken mode (i.e., nucleation mode) particles with large aerosol than for coagulation of two Aitken mode particles. Coagulation is also enhanced in shearing or turbulent flows, as these induce fast relative particle motion.

Condensation. As long as the partial pressure of a compound in the gas phase is higher than the vapor pressure of that compound in aerosol, growth will occur. The opposite situation is possible, where particles outgas certain compounds, but the growth tends to be the dominant process. Condensational growth is another mechanism by which aerosol can leave the nucleation mode.

Cloud processing. Cloud drops undergo aqueous chemical reactions. Because of their larger surface area, reaction rate per drop is usually higher than reaction rate per condensation nucleus. If a drop that has been involved in such reactions subsequently evaporates, the condensation nucleus is left behind and may be considerably larger than it was before entering cloud. This process is called cloud processing, and can considerably increase the rate at which accumulation mode aerosol grow. Cloud processing generally affects accumulation mode rather than coarse mode aerosol, as the former tends to be more hygroscopic.

Four major processes by which clouds can impact aerosol populations:

• Vertical transport

• Scavenging processes (both with water and ice hydrometeors)

• Chemical processes in cloud drops

• Particle formation near clouds

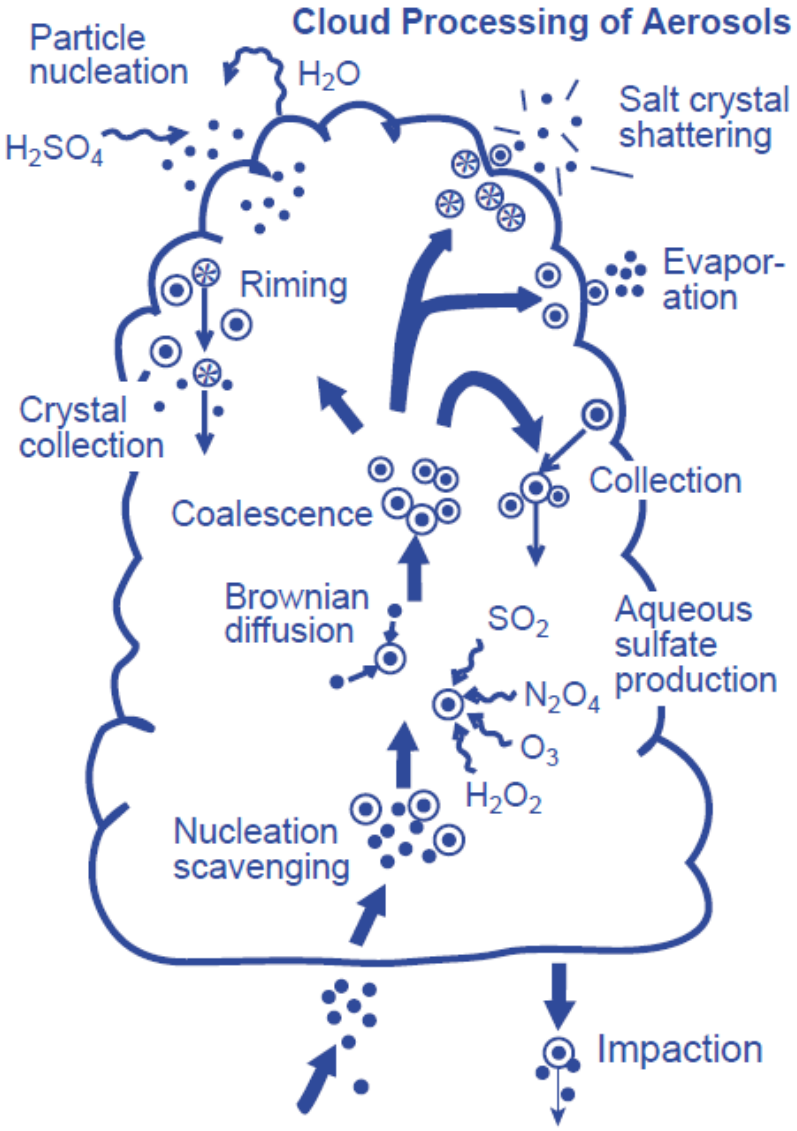


Figure 5.4 A schematic diagram of the in- or near-cloud processes that can alter the aerosol size distribution

An idealized cloud:

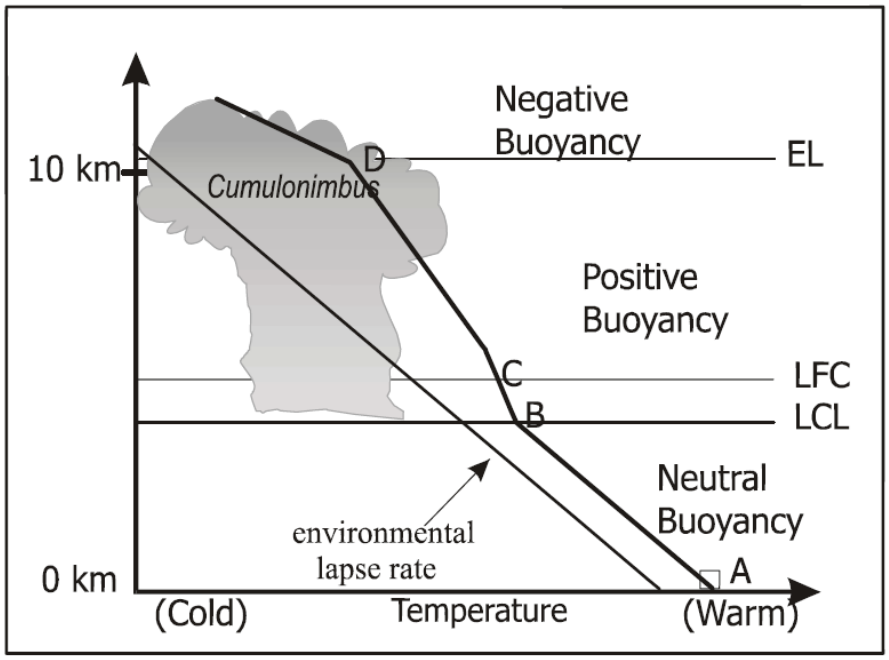


Figure 5.5 Temperature profile of idealized cloud.

- Between points A and B, the parcel cools at the dry adiabatic lapse rate (-9.8^{o}C km^{-1}).
- At B the parcel becomes saturated and condensation initiates (called Lifting Condensation Level (LCL), and varies in height according to the initial temperature and moisture content of the parcel).
- Once water droplets or ice crystals form, latent heat (energy released/required in phase changes) is released into the parcel, increasing its temperature. Further cooling due to lifting occurs at the moist adiabatic lapse rate which varies between 4^{o}C km^{-1} in the lower troposphere to 7^{o}C km^{-1} in the middle and upper troposphere.
- If the lifting continues past the LCL, it is possible that the cloud reaches a elevation where it becomes positively buoyant (point C) (called the Level of Free Convection (LFC).
- Once the top of a cloud passes the LFC, it continues to rise until it either runs out of moisture or when it encounters a stable layer of warm air. The height at which the parcel is no longer positively buoyant is called the equilibrium height (point D). The height of the LFC is a function of environmental lapse rate. Cool or cold air in the mid-troposphere usually has a high environmental lapse rate (>7^{o}C km^{-1}), which is very conducive to cumulus development. The closer the LFC is to the LCL, the easier it is for cumulus clouds to develop.
- *In the simplest of terms, clouds that are contained between the LCL and LFC are stratiform while those that continue to grow above the LFC are cumuliform.*

Major cloud processes.

Microphysics of warm clouds:

- ✓ *Nucleation of drops*

  Homogeneous nucleation is not efficient, requires supersaturation of 300-400%. Heterogeneous nucleation requires cloud condensation nuclei (CCN).

- ✓ *Condensation and evaporation*

  Once formed, water drops may continue to grow as vapor diffuses toward them (this process is called condensation). The reverse process, drops decreasing in size as vapor diffuses away from them, is called evaporation.

- ✓ *Fallout of drops*

  Depends on drop size. Terminal fall speed is when gravitational force is balanced by friction.

- ✓ *Coalescence*

  Continuous collection: cloud drop growth by coalescence with other drops as a drop falling through a cloud;

  Stochastic collection: cloud drop growth in a discrete, stepwise, probabilistic manner.

- ✓ *Breakup of drops*

  When raindrops achieve a certain size, they become unstable and break up into smaller drops.

Microphysics of cold clouds:

- ✓ *Homogeneous nucleation of ice particles*

  Nucleation of ice particles from either liquid or water vapor phase. Occurs at lower than about -35 to -40^0C (unfrozen water is called supercooled but freezes spontaneously below -40^0C)

- ✓ *Heterogeneous nucleation of ice particles*

  Several modes: condensational nucleation, immersion freezing, deposition nucleation, and contact nucleation. Occurs at temperature from 0C to -40^0C.

- ✓ *Deposition and sublimation*

Growth of in ice particle by diffusion of ambient vapor toward the particle is called deposition. The lost of mass of an ice particle by diffusion of vapor from the ice-phase into the air is called sublimation.

- ✓ *Aggregation and riming*

  If ice particle collect other ice particles, the process is called aggregation. If ice particles collect liquid drops, which freeze on contact, the process is called riming.

  Extreme riming produces hail (~ 1 cm, can be as large as 10-15 cm).

- ✓ *Ice enhancement*

  Far more ice particles than ice nuclei. Hypotheses: 1) fragmentation of ice crystals; 2) ice splinter production in riming

- ✓ *Fallout of ice particles*
- ✓ *Melting*

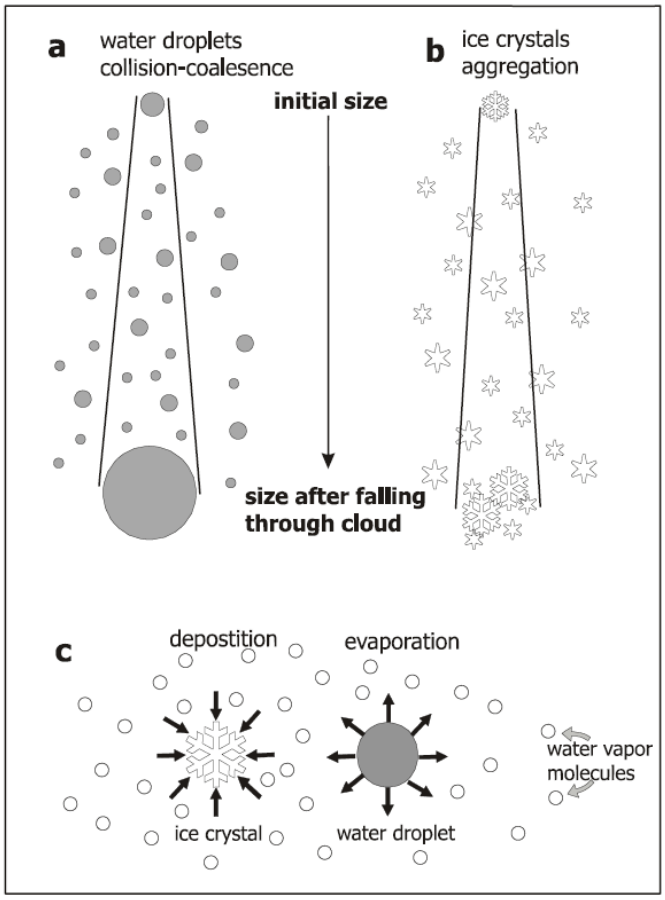


Figure 5.6 Growth of precipitation.

Growth in warm (liquid) clouds:

- ✓ Small droplets evaporate, producing extra water vapor which is in turn made available to the larger droplets. Once this process is initiated, a select few droplets grow quite rapidly. When droplets reach a size of about d= 0.1 mm, they start to fall through the cloud.
- ✓ Falling large collide and coalesce (merge) with the smaller droplets in a process called collision-coalescence (Figure 5.6a). Large droplets exit the base of the cloud as soon as their fall velocities exceed the speed of any updrafts.

Growth in mixed phase (liquid-ice) clouds:

- ➢ Ice crystals can grow at the expense of water droplets. When this occurs some of the ice crystals grow large enough to fall through the cloud, colliding and merging with other ice crystals, in a process called aggregation (Figure 5.6b)

Ice formation processes distinguishing ice nucleation occurring on aerosol particles from other processes affecting cloud particle composition, such as the formation of ice from secondary processes involving the collision of preexisting ice crystals, the splintering of ice off of graupel during the accretion of cloud droplets, and the fracture of ice crystals exposed to dry air (from DeMott et al., BAMS, 2011)