Aerosols

Modifications of elementary parcel theory

Account for the burden of condensed water

- If condensed water is present in the parcel, it exerts a downward force on the parcel equal to its weight.
- The buoyancy factor B then becomes:

$$B = \frac{\rho^i - \rho}{\rho} = \frac{\frac{p}{RT'} - \left(\frac{p}{RT} + \rho_l\right)}{\frac{p}{RT}} = \frac{T}{T'} - \left(1 + \frac{\rho_l}{\rho}\right) = \frac{T}{T'} - (1 + \mu) \quad (1)$$

where μ [kg condensate/kg air] = mixing ratio of the condensate

Compensating downward motion

- The mass flux (MF) of upward-moving air through the level is ρUA [kg/s] and the downward MF is $\rho' U' A'$ (U = velocity of thermals).
- If the area of consideration is large enough then: $MF_u = MF_d$ or:

$$\frac{A}{A'} = \frac{\rho' U'}{\rho U} \sim \frac{U'}{U} \quad (2)$$

assuming $\rho' \sim \rho$.

- $\Rightarrow$ smaller updrafts have higher velocities
- Further assume that ascending air follows pseudoadiabatic lapse rate, while descending air follows dry adiabatic lapse rate.
- Thus, after a short time dt, the air arriving from level below will have a temperature given by $T_o + (\gamma - \Gamma_s)$ U dt, where T_o is the initial temperature at that level. Γ_S denotes the pseudoadiabatic lapse and γ the ambient lapse rate.

- Air arriving from above has temperature: $T_o + (\Gamma - \gamma)\, U' \, dt$.
- The situation is unstable when this temperature is less than the temperature of the thermal, i.e.:

$$(\gamma - \Gamma_s)U > (\Gamma - \gamma)U' \quad (3)$$
$$\leftrightarrow (\gamma - \Gamma_s)A' > (\Gamma - \gamma)A \quad (4)$$

- in the limit as A goes to zero, this is equivalent to $\gamma > \Gamma_s$ (unstable).
- The neutral case arises when

$$\frac{\gamma - \Gamma_s}{\Gamma - \gamma} = \frac{A}{A'} \quad (5)$$

- if $\frac{A}{A'} > 0$ (that is thermals are not negligible in size) this equation can only be satisfied if $\gamma > \Gamma_s$ (what was unstable before is now neutral).
- $\Rightarrow$ the ambient lapse rate must be steeper for instabilities to occur when compensating downward motions are taken into account

Dilution by mixing

- When a buoyant element ascends, some mixing takes place through the boundaries. Since the ambient air is generally cooler and drier than the buoyant element, mixing will reduce both the buoyancy of the thermal and lower its mixing ratio (entrainment).
- Account for entrainment by considering heat exchange between cloudy air and entrained air.
- Denote m as mass of cloudy air, which consists of dry air, water vapor and condensed water. Entrain mass dm through lateral sides as the cloud ascends through height dz.
- Heat required to warm the entrained air is:

$$dQ_1 = c_p(T - T')dm \quad (6)$$

where temperature of cloudy air $= T$, of ambient air $= T'$ and the heat contents of the vapor and the condensate are neglected.

- Next assume that just enough condensate evaporates to saturate the mixture. The heat required is:

$$dQ_2 = L(w_s - w')dm \quad (7)$$

where w' is the mixing ratio of entrained air.

- Condensation occurs during the ascent, gaining latent heat:

$$dQ_3 = -mL dw_s \quad (8)$$

i.e. the parcel loses heats $dQ_1 + dQ_2$ but gains dQ_3.

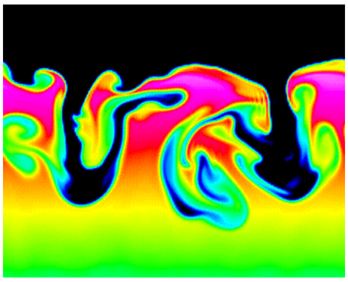
- From 1. law of thermodyn:

$$m(c_p dT - R_d T\frac{dp}{p}) = -dQ_1 - dQ_2 + dQ_3 \quad (9)$$

- divide both sides by mc_pT:

$$\left(\frac{dT}{T} - \frac{R_d}{c_p}\frac{dp}{p}\right) = \frac{-dQ_1 - dQ_2 + dQ_3}{mc_pT} \quad (10)$$
$$\frac{d\Theta}{\Theta} = -\frac{L}{c_pT}dw_s - \left[B + \frac{L}{c_pT}(w_s - w')\right]\frac{dm}{m} \quad (11)$$

- without entrainment (dm = 0) we get back the change in Θ due to the pseudoadiabatic process.
- Because the bracketed term is always positive in cases of interest, the above equation implies that the temperature falls off at a faster rate with entrainment, i.e. buoyancy is impaired by entrainment.
- Alternative to lateral mixing is mixing of dry environmental air from just above cloud top.
- Turbulence draws parcel of ambient air into the cloud, causing the evaporation of some cloud droplets. This will chill the parcel, reduce its buoyancy and lead to a downdraft.
- The cumulative effect of many such penetrative downdrafts from cloud top will be to cool and dry the cloud, especially in its upper regions.
- It is said to be the mechanism for the break-up of stratus clouds into stratocumulus when going from subtropics to tropics.



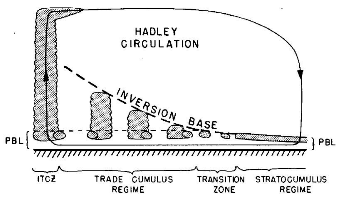
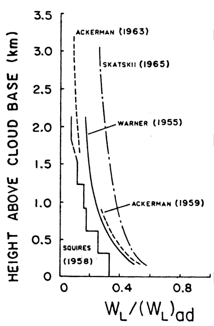


FIG. 5. A schematic illustration of the role of CIFKU in determining the tropical and subtropical distributions of cloudiness. Details are given in the text.

adiabatic liquid water content (LWC)



- In most cases LWC < LWC_{ad} → parcel theory
- LWC ~ LWC_{ad}:
  1) near cloud base
  2) within undiluted "adiabatic" pockets
- LWC/LWC_{ad} ~ 0.1 and 0.6

Fig. 2-22: Ratio of the observed mean liquid water content at a given height above cloud base to the adiabatic value, for non-precipitating clouds. (From Warner, 1970a; by courtesy of Am. Meteor. Soc., and the author.)

→ cloud as a whole has significantly less water than adiabatic value, because of entrainment

Aerodynamic resistance: thermals and plumes

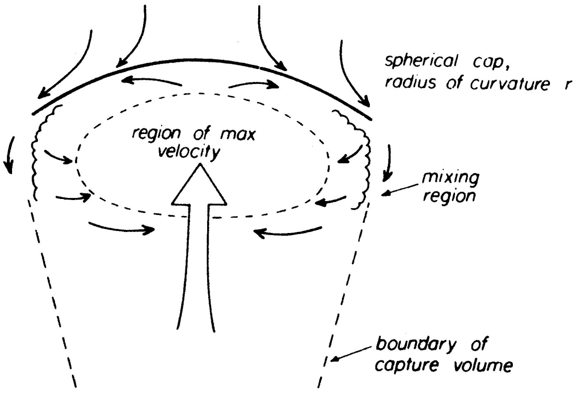


FIG. 4.3. Structure of a convective bubble.

- This is an idealized thermal (spherical cap, radius of curvature r), based on laboratory studies
- Resembles atmospheric thermals which appear as "turrets" or protuberances of cumuli. They are said to be shape-preserving.
- Thus, the vertical velocity of a bubble depends on its size and buoyancy according to:

$$u = c\sqrt{g\overline{B}r} \tag{12}$$

  where u is the upward velocity of the cap, $\overline{B}$ is the average buoyancy factor across the bubble, r is the radius of the cap, and c=1.2 is given from experiments
- In the atmosphere, however, cumuli are more complicated than these elementary bubbles.
- Their velocity is related to the stability of the air and the size and state of development of the cloud as a whole, and cannot be predicted for all clouds and for all occasions with the above equation.

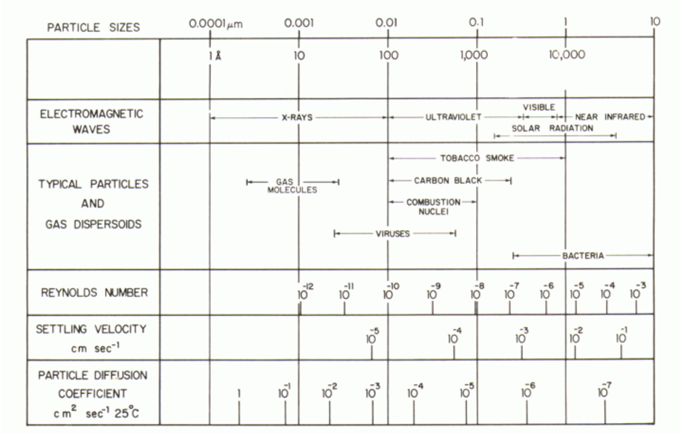
Aerosols: Definition

- Definition of an aerosol: disperse system with air as carrier gas and a solid or liquid or a mixture of both as disperse phases.
- Aerosol particles (AP) are in the radius range from $10^{-3}\mu$m to several hundred μm. They are larger than atmospheric small ions:
-

	diameter (μm)	mass (g)	concentration (cm^{-3})
N_2	0.00038	$4.6 \cdot 10^{-23}$	10^{19}
AP	0.01 - 10	10^{-18} - 10^{-9}	$< 10^8$

- nucleation mode :$10^{-4}\mu$m - $10^{-1}\mu$m
- accumulation mode: $10^{-4}\mu$m - 1 μm
- coarse mode AP: $> 1\mu$m

Size classes of aerosol particles



Aerosol processes (Fig. 2.15 Seinfeld&Pandis Figure)

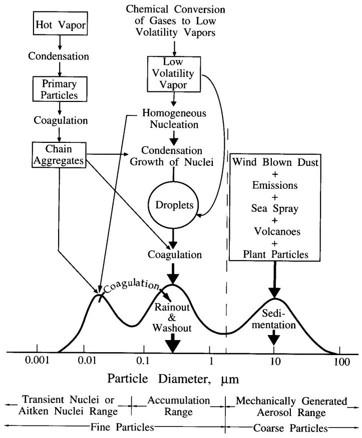
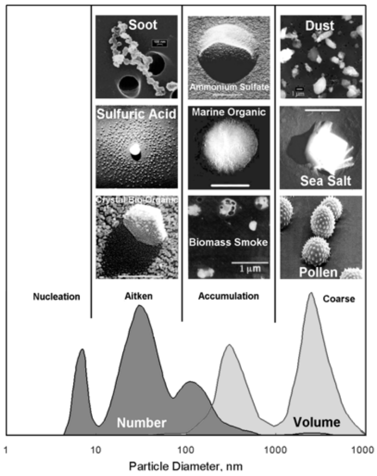


FIGURE 2.15 Idealized schematic of the distribution of particle surface area of an atmospheric aerosol (Whitby and Cantrell, 1976). Principal modes, sources, and particle formation and removal mechanisms are indicated.

Aerosol processes in the different modes

- For AP > 1 μm gravitational settling (or dry fallout) is important as a removal mechanism (1 μm AP $\sim$ 0.003 cm/s ; 10 μm AP $\sim$ 0.3 cm/s)
- Anthropogenic AP are mainly in submicron range; natural aerosols are mainly in supermicron range
- Nucleation mode AP originate primarily from combustion processes, of which the most important sources are probably related to human activities (but forest fires and volcanoes also contribute)
- Coagulation of nucleation mode AP is major source for large AP. Coagulation does not remove AP from the atmosphere, it modifies their size spectra and shifts nucleation mode nuclei into size ranges where they can be removed by other mechanisms
- Coarse mode peak attributable to mechanical processes, such as wind erosion which produces dust
- Mobility of AP decreases rapidly as their size increases, i. e. coagulation due to Brownian motion is essentially confined to AP < 0.2 μm

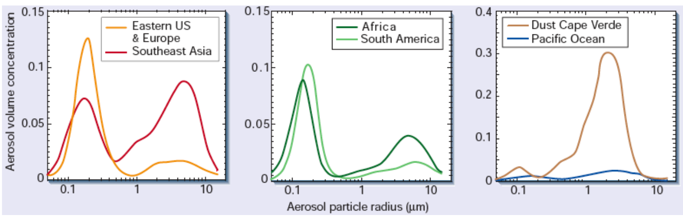
Different aerosol types (Source: IGAC, 2003)



Aerosol size distributions (Kaufman et al., 2002)

Atmospheric aerosol size distribution is usually presented as a sum of n log-normal distributions because D_p typically varies over several orders of magnitude:

$$n(Dp) = \sum_{i=1}^{n} \frac{N_i}{\sqrt{2\pi} D_p ln\sigma_i} exp\left[\frac{-(lnD_p - ln\overline{D}_{pi})^2}{2ln^2\sigma_i}\right] \qquad (13)$$



Aerosol size distributions (Kaufman et al., 2002)

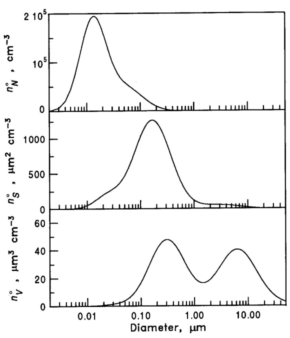


FIGURE 7.12 Typical urban aerosol number, surface, and volume distributions.

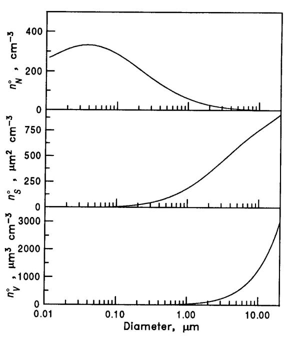


FIGURE 7.22 Typical desert aerosol number, surface, and volume distributions.

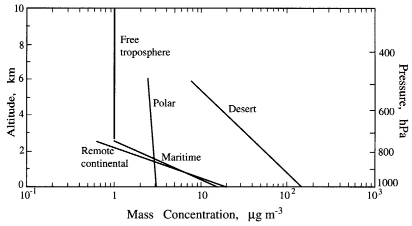


FIGURE 7.25 Representative vertical distribution of aerosol mass concentration (Jaenicke, 1993).

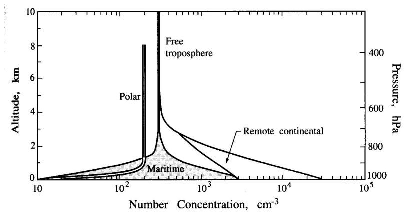


FIGURE 7.26 Representative vertical distribution of aerosol number concentration (Jaenicke, 1993). A range of concentrations is shown for marine and remote continental aerosols.

Source: Figures 7.25 & 7.26 Seinfeld&Pandis

Formation of aerosols

- Gas-to-particle conversion: nucleation of AP from supersaturated gases
- Bulk-to-particle conversion: wind blown dust (arid regions), emissions of pollens and spores by plants, and over oceans
- Liquid-to-particle conversion: coarse mode AP composed of sea-salt originate from drops ejected into the air when air bubbles in breaking waves burst at ocean surface.

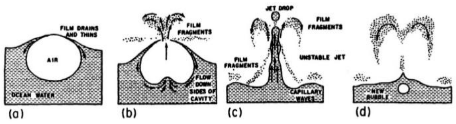


Fig. 8-12: Four stages in the production of sea salt particles by the bubble-burst mechanism. (a) Film cap protrudes from the ocean surface and begins to thin. (b) Flow down the sides of the cavity thins the film which eventually ruptures into many small fragments. (c) Unstable jet breaks into few drops. (d) Tiny salt particles remain as drops evaporate; new bubble is formed. (From Day, 1965, with changes.)

-

Global Source Strength, Lifetime and Burden

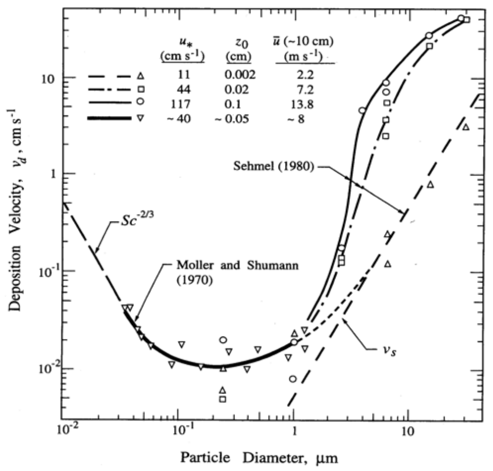
		Aerosol Type	Flux (Tg/yr)	Lifetime (d)	Burden (mg/m^2)
Natu-ral	Pri-mary		900-1500	4	19-33
			2300	1	3
			50	4	1
	Sec.		70	5	2
			20	10	1
			(40)	(400)	(80)
			20	5	0.6
	Total		3400-4000		27-41
Anth-ropo-genic	Pri.		40-640	4	1-14
			14	7	0.6
			54	6	1.8
	Sec.		140	5	3.8
			20	7	0.8
	Total		270-870		8-21
Sum			3670-4870		35-62

(Source: Ramanathan et al., Science, 2001)

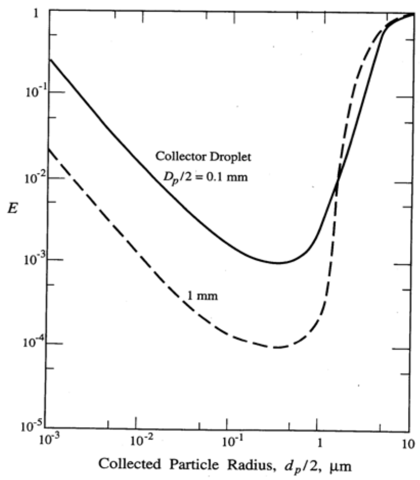
Removal of aerosols

- 80%-90% of the AP mass is removed from the atmosphere by precipitation particles (wet scavenging)
- prior to that AP serve as nuclei upon which cloud particles form. As these particles grow, AP tend to be forced onto their surface by diffusion fields associated with the flux of water vapour to the growing CD (diffusiophoretic force)
- AP < 0.1 μm are collected most efficiently by diffusiophoresis
- precipitation particles collect AP by direct impaction, the better the larger the AP (best for AP > 2 μm).
- AP are also removed by gravitational settling and subsequent impaction onto obstacles on Earth's surface (dry deposition) which accounts for 10-20% of AP mass removed from the atmosphere.

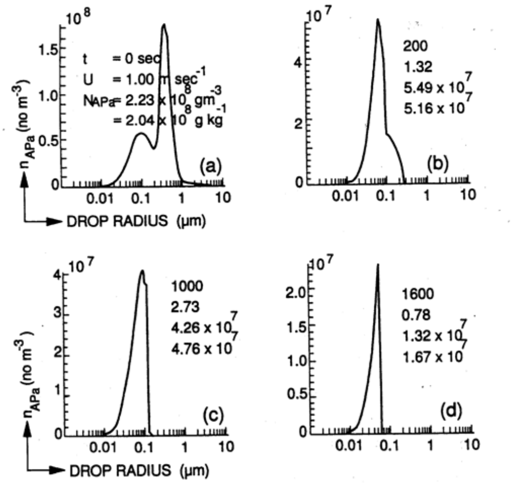
Dry deposition (Fig 19.3 Seinfeld&Pandis)



Wet deposition (Fig 20.10 Seinfeld&Pandis)



Nucleation scavenging (Fig 17.8 Pruppacher&Klett)



Residence time of aerosols (Fig 8.14 Pruppacher&Klett)

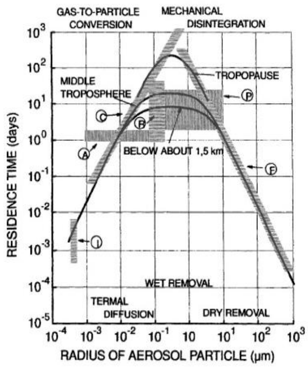


Fig. 8-14: Residence time of aerosol particles as a function of their radius. I: Small ions, A: Aitken particles, C: from thermal diffusion of aerosol particles, R: based on radioactivity data, P: removal by precipitation, F: removal by sedimentation. (From Jaenicke, 1978a, 1988, with changes.)

Residence time of aerosols (Fig 8.15 Pruppacher&Klett)

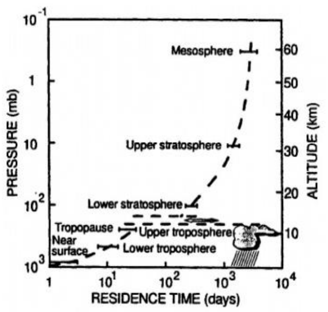
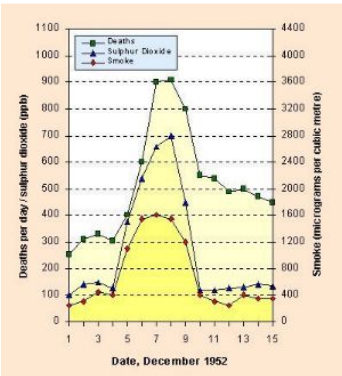


Fig. 8-15: Residence time of aerosol particles as a function of altitude in the atmosphere. (From Jaenicke, 1978c, 1988, based on Flohn, 1973, with changes.)

Importance of aerosols

- Aerosol particles act as centers for cloud droplets and ice particles
- Effects on pollution:
  - photochemical smog (ozone)
  - degradation of visibility
  - winter smog (solid aerosols provide surface upon which trace gases can be absorbed and then react, e.g. *London smog*)
- Effects on climate - effects on radiative transfer (direct and indirect effect)

London smog



Aerosol radiative effects