Particles and Scattering

Topics:
1. Particle size distributions
2. Single scattering albedo/phase functions
3. Cross sections and efficiencies
4. Polarization of radiation
5. Rayleigh scattering
6. Index of refraction

Reading: Liou 3.3.1; Thomas 3.4, 9.3

Aerosols

Small particles ($< 10\ \mu$m), solid or liquid
1. Sulfates - (sulfuric acid, ammonium sulfate) hydrated depending on RH.
From gas-to-particle conversion of SO_2, DMS.
2. Mineral - wind blown dust from soils (quartz, iron oxide) (large size).
3. Sea salt - from air bubbles bursting at sea surface (large).
4. Combustion - from fossil fuels, forest burning (small).

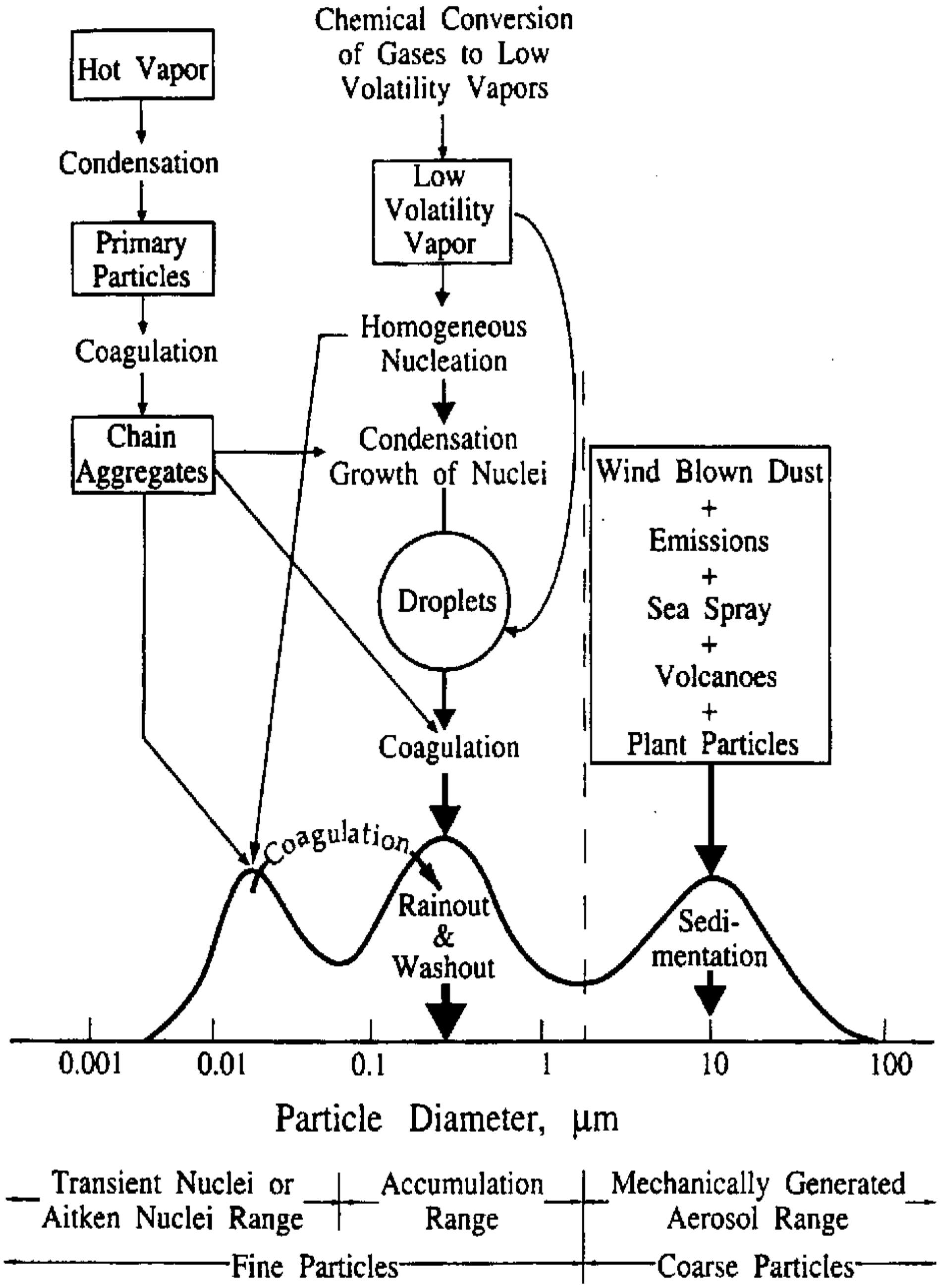
Aitken nuclei (nucleation mode) $< 0.1\ \mu$m
large aerosol (accumulation mode) 0.1 to $2.0\ \mu$m (most important for radiation)
giant aerosol (coarse mode) $> 2.0\ \mu$m

Clouds

Types: cumulonimbus - deep thunderstorms;
cirrus - high altitude (cold) ice clouds;
altostratus, altocumulus - midlevel supercooled liquid clouds;
stratus and cumulus - low (boundary layer) liquid clouds.

Cloud microphysics:
Water droplets - radius: 2 - 40 μm radius (typical radius 10 μm)
Concentration: 50 - 500 cm^{-3}
Liquid water content: 0.03 - 3.0 g/m^3

Ice clouds - non-spherical particles
Size: length 10 - 1000 μm, typical length 100 μm
Concentration: 1 - 1000 liter^{-1}
Ice water content: 0.001 - 0.1 g/m^3



Idealized schematic of the particle surface area distribution of atmospheric aerosols. [from Whitby and Cantrell, 1976]

Particle Size Distributions

Interaction of radiation with particles depends on radius (and composition).

Size distribution $n(r)$ is the number of particles per volume per radius interval.
Typical units: $\mathrm{cm}^{-3}\ \mu\mathrm{m}^{-1}$.
Total number of particles per volume is integral

$$N = \int n(r)dr$$

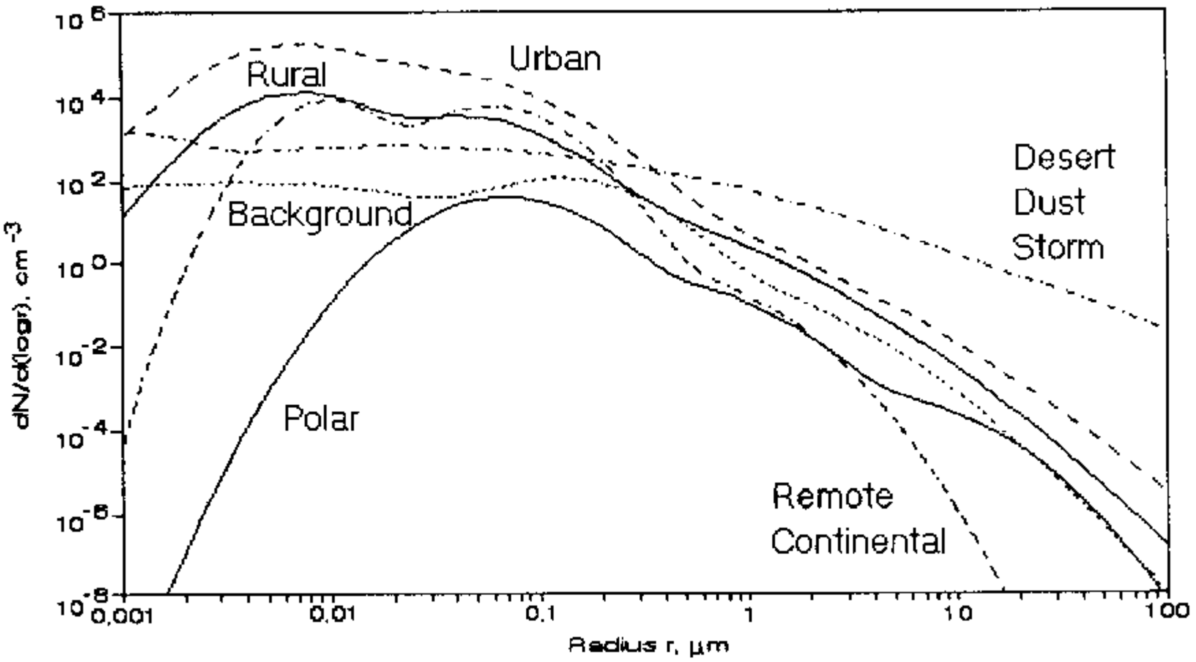
Mass per volume of air or liquid water content (LWC) is

$$W = \frac{4\pi}{3}\rho_l \int r^3 n(r)dr$$

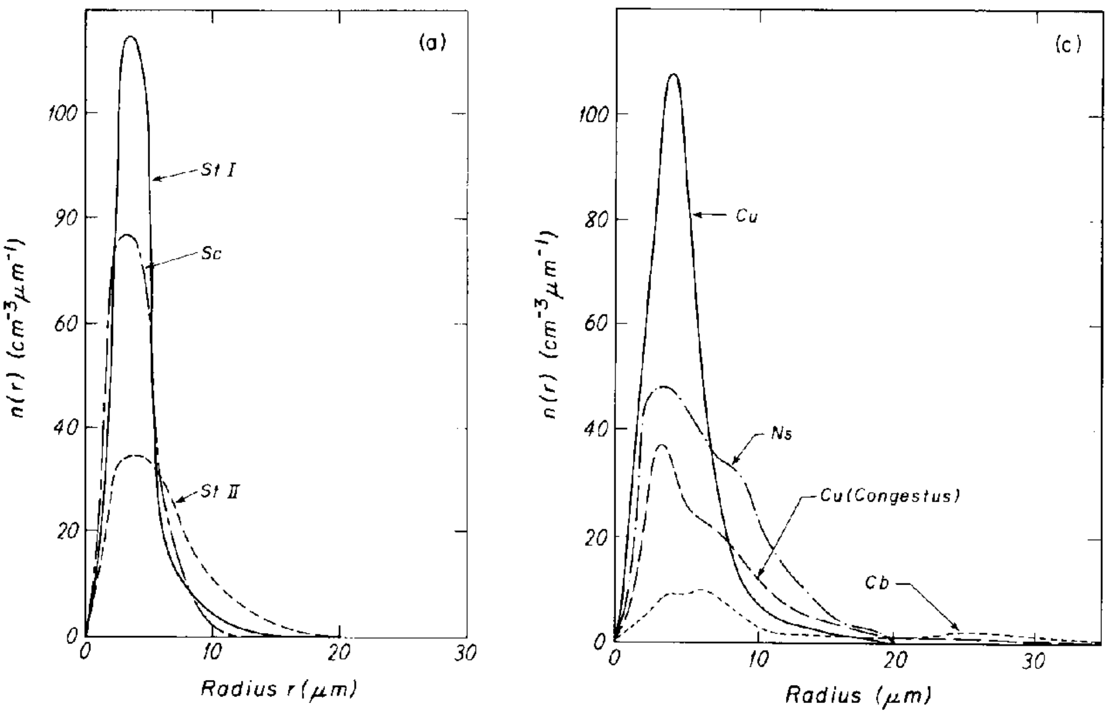
ρ_l is density of particle material (e.g. water).
Effective radius is ratio of third and second moments:

$$r_{\mathrm{eff}} = \frac{\int r^3 n(r)dr}{\int r^2 n(r)dr}$$

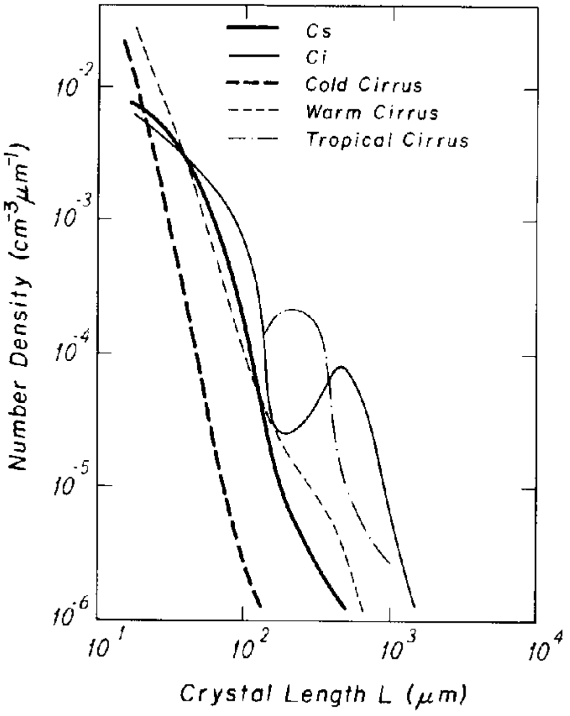
In aerosol research the surface area distribution ($\mu\mathrm{m}^2\ \mathrm{cm}^{-3}\mu\mathrm{m}^{-1}$) or the volume distribution ($\mu\mathrm{m}^3\ \mathrm{cm}^{-3}\mu\mathrm{m}^{-1}$) are also used.



Representative aerosol size distributions. [R. Jaenicke, 1993]



Representative liquid cloud droplet size distributions. [Liou, 1992]



Representative ice cloud crystal size distributions. [Liou, 1992]

Theoretical size distributions

Gamma and lognormal distributions are used to represent particle size distributions. Aerosol size distributions often represented by sum of three lognormal distributions (which characterize the three production modes).

The gamma distribution is

$$n(r) = \frac{Nb^{\alpha+1}}{\Gamma(\alpha+1)} r^{\alpha} e^{-br}$$

where $r_c = \alpha/b$ is the modal radius and α controls the width.
Moments of the gamma size distribution are

$$\int_0^{\infty} r^k n(r) dr = \frac{Nb^{-k}\Gamma(\alpha+k+1)}{\Gamma(\alpha+1)}$$

Effective radius is $r_{\text{eff}} = (\alpha+3)/b$; effective variance is $v_{\text{eff}} = 1/(\alpha+3)$.
Mass content is $W = \frac{4\pi}{3}\rho_l N b^{-3}(\alpha+3)(\alpha+2)(\alpha+1)$.

The log-normal distribution is

$$n(r) = \frac{N}{\sigma\sqrt{2\pi}} \frac{1}{r} \exp\left[-\frac{(\ln(r/r_0))^2}{2\sigma^2}\right]$$

r_0 is the modal radius (in $\ln r$), σ is the standard deviation of $\ln r$.
Note: Often σ is replaced by $\ln \sigma$.

Moments of lognormal size distribution are

$$\int_0^{\infty} r^k n(r) dr = N r_0^k \exp(k^2\sigma^2/2)$$

Effective radius is $r_{\text{eff}} = r_0 \exp(2.5\sigma^2)$; effective variance is $v_{\text{eff}} = e^{\sigma^2} - 1$.
Mass content is $W = \frac{4\pi}{3}\rho_l N r_0^3 \exp(4.5\sigma^2)$.

Light Scattering by Particles

Scattering is the redirection of light.
Radiation can be attenuated by absorption or scattering or both.

Amount of scattering by a distribution of particles is usually quantified with the volume scattering coefficient:
β_{sca} = fraction of radiation scattered per distance beam travels.

Particles attenuate by both absorption and scattering: $\beta_{ext} = \beta_{abs} + \beta_{sca}$

The relative importance of absorption and scattering is given by the
single scattering albedo: $\omega = \beta_{sca}/\beta_{ext}$.
No scattering $\omega = 0$. No absorption $\omega = 1$ (conservative scattering).

Phase Functions

The directional distribution of scattered radiation is the *phase function*:
$P(\Theta)$ is the fraction scattered in direction Θ.

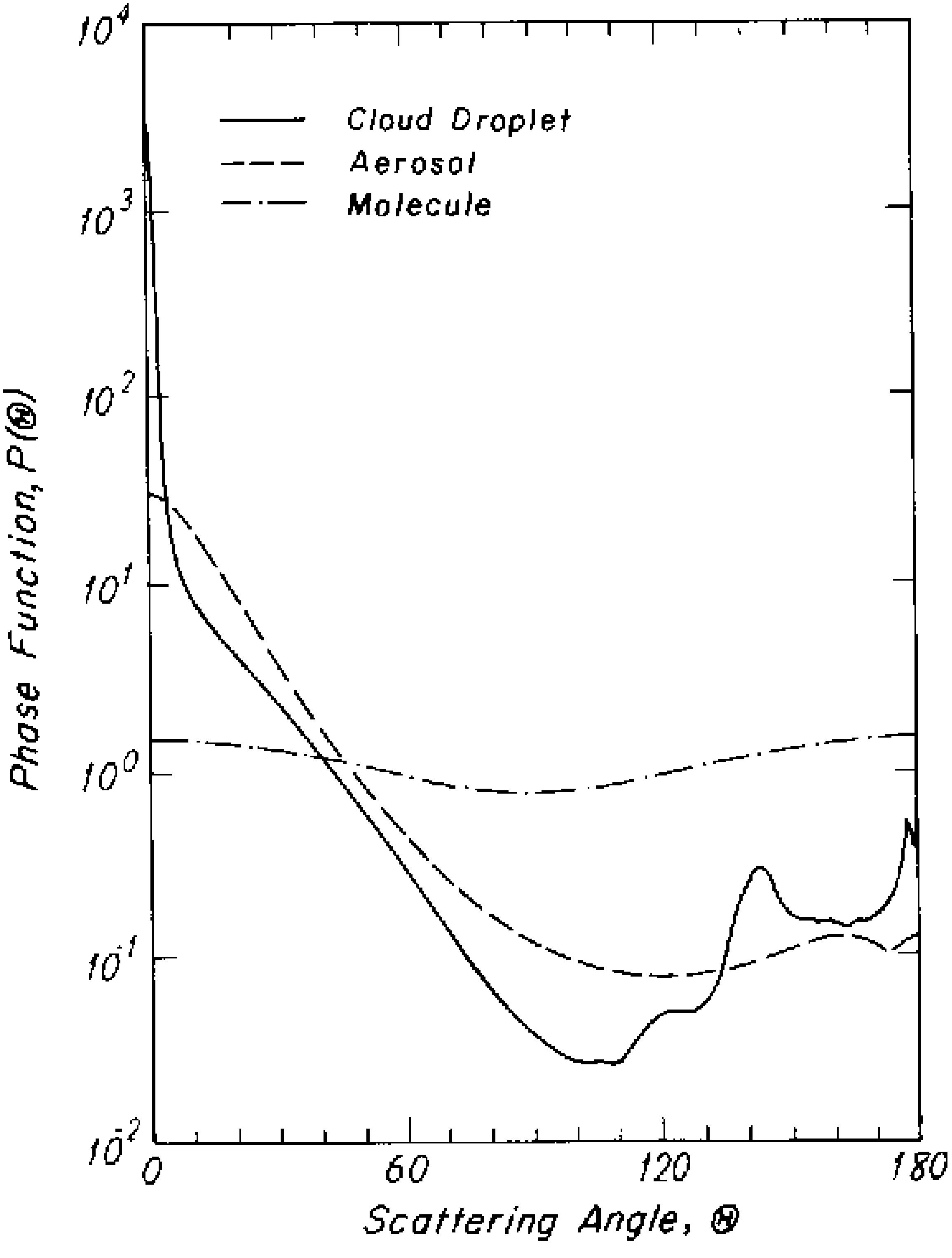
Θ is the scattering angle:
$\Theta = 0$ is forward scattering, $\Theta = 180°$ is backscattering.

Normalization: $\int_0^{2\pi} \int_0^{\pi} P(\Theta)\ d(\cos\Theta)\ d\phi\ =\ 4\pi$
Isotropic scattering (equal in all directions) has $P(\Theta) = 1$.

Asymmetry parameter is first moment of phase function:

$$g = \frac{1}{2} \int_{-1}^{1} P(\cos\Theta)\ \cos\Theta\ d(\cos\Theta)$$

$g = 0$ for equal forward and backward. $g = 1$ for totally forward.
For water clouds in visible $g \approx 0.85$.



Example phase functions for cloud droplets, aerosols, and molecules for a wavelength of 0.5 μm. [Liou, 1992, Fig. 3.5]

Single Particle Scattering

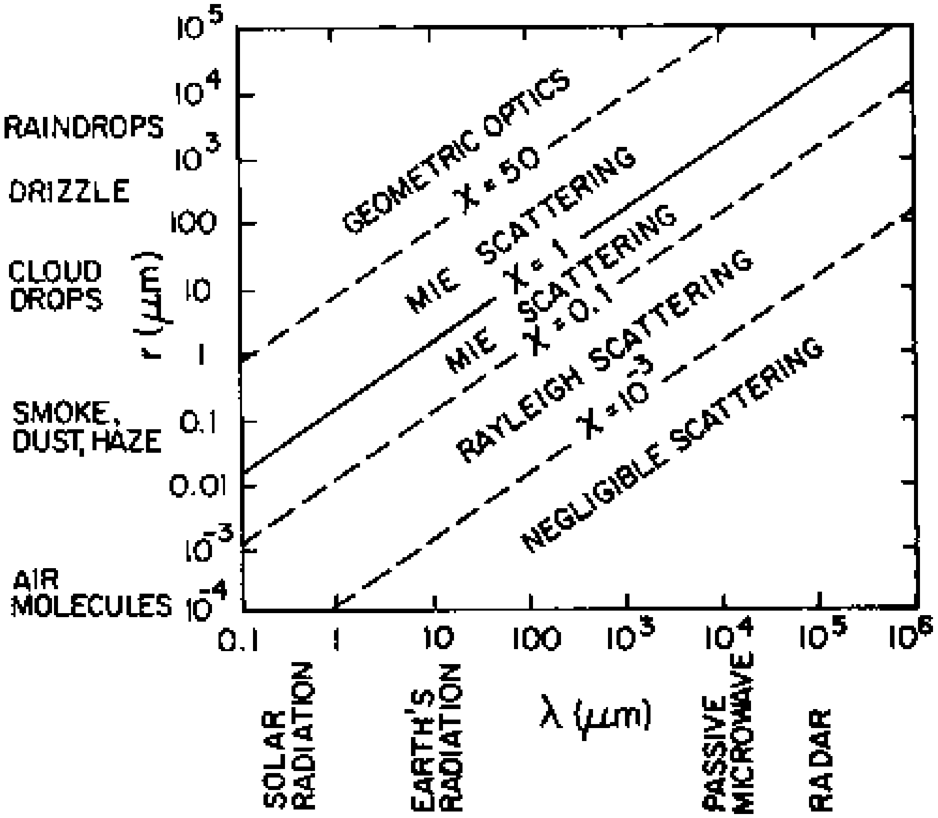
Spherical particle scattering depends on:

1) index of refraction m
2) size parameter x

$$x = \frac{2\pi r}{\lambda}$$

r = particle radius, λ = wavelength.

Scattering characteristics depend strongly on size parameter.



The scattering regime depends on size parameter $x = 2\pi r/\lambda$. [Wallace and Hobbs, 1977]

Extinction, scattering, and absorption for single particles is measured in cross sectional area, C_{ext}, C_{sca}, C_{abs}.

Volume extinction coefficient related to cross section by $\beta_{ext} = NC_{ext}$ (N is number of particles per volume).

Efficiency factors: unitless ratio of cross section to projected area of particle:

$$Q_{ext} = \frac{C_{ext}}{\pi r^2} \qquad Q_{sca} = \frac{C_{sca}}{\pi r^2}$$

Scattering from Particle Distributions

Integrate over particle size distributions and multiple particle types.

Scattering properties needed for radiative transfer:

1) Extinction

$$\beta_{ext} = \int_0^\infty C_{ext}\, n(r) dr \ = \int_0^\infty \pi r^2 Q_{ext}\, n(r) dr$$

2) Single scattering albedo

$$\omega = \frac{\beta_{sca}}{\beta_{ext}} \ = \frac{\int_0^\infty C_{sca}\, n(r) dr}{\int_0^\infty C_{ext}\, n(r) dr}$$

3) Phase function or asymmetry parameter g

$$P(\Theta) = \frac{\int_0^\infty P(\Theta) C_{sca}\, n(r) dr}{\int_0^\infty C_{sca}\, n(r) dr}$$

Do *not* add ratios like single scattering albedo or phase function.
Instead add quantities $\propto n(r)$, e.g. $\tau\omega g = \tau_1\omega_1 g_1 + \tau_2\omega_2 g_2$

Polarization of Radiation

The electric field is a vector (perpendicular to propagation direction).

Polarized nature of light is required to understand particle scattering.

Polarization is defined relative to a reference plane. Two components of electric field ($\hat{||}$ is parallel to plane, $\hat{\perp}$ is perpendicular):

$$\mathbf{E} = E_{||}\hat{||} + E_{\perp}\hat{\perp}$$

Each component is an oscillating electric field with amplitude $|E_0|$ and phase $\arg E_0$ (E_0 is complex amplitude):

$$E_{||} = E_{0,||} \exp[ik(x - ct)] \qquad E_{\perp} = E_{0,\perp} \exp[ik(x - ct)]$$

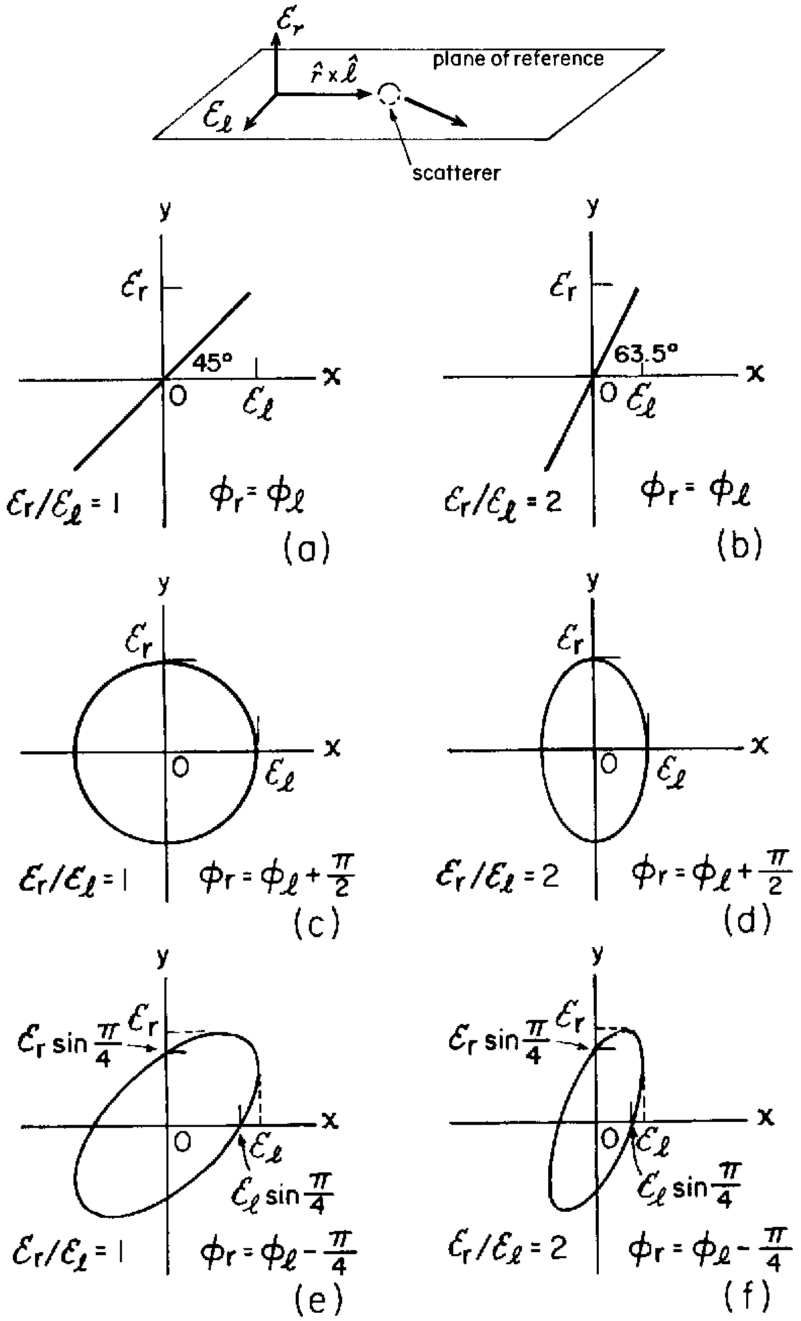


Illustration of the patterns the E field vector traces out. The E field has components in the parallel (ℓ) and perpendicular (r) directions. The type of polarization is determined by the relative amplitudes ($\mathcal{E}$) and phases (ϕ) of the two components. [Stephens, 1994; Fig. 2.9]

Tip of E field vector - $E_\perp$ vs. $E_{||}$:

linear polarization: $\Phi_{||} = \Phi_\perp$

circular polarization: $\Phi_{||} = \Phi_\perp + \pi/2$, $E_{0,\perp} = E_{0,||}$

Single EM wave specified by $E_{||}$, $E_\perp$, and $\Phi_{||} - \Phi_\perp$.

Actual light is a superposition of **many waves** of different frequencies, phases, and amplitudes.
Polarization is determined by the relative size and correlations between the two electric field components.
Unpolarized - even mixture, no correlations (sunlight, most emission).

Stokes Parameters

Four Stokes parameters specify the state of polarization:

$$I_{||} = \langle |E_{||}|^2 \rangle \quad I_{\perp} = \langle |E_{\perp}|^2 \rangle \quad \text{or} \quad I = I_{||} + I_{\perp} \quad Q = I_{||} - I_{\perp}$$

$$U = \langle E_{||} E_{\perp}^* + E_{\perp} E_{||}^* \rangle \quad V = i \langle E_{||} E_{\perp}^* - E_{\perp} E_{||}^* \rangle$$

$\langle\rangle$ means time average (or incoherent sum over single waves).
Stokes parameter have units of intensity; I is radiance.
Q and U require a reference plane, usually the scattering plane.

I is the sum of power in each component ($I_{||}$ and $I_{\perp}$).
Q is difference between the power in the two components.
U is correlation between the $||$ and $\perp$ fields; max U for linear polarization at $45°$.
V is correlation of $||$ with $\perp$ shifted $90°$ in phase.

Vertical and horizontal polarization:

$$I_V = I_{||} = (I + Q)/2 \quad I_H = I_{\perp} = (I - Q)/2$$

Radiation may be unpolarized, partially polarized, or fully polarized.

Unpolarized: $Q = U = V = 0$.
Unpolarized light is made of many independent waves; mean intensity is $I/2$ for each component; random phases imply no correlation so $U = 0$ and $Q = 0$.
Fully polarized: $I^2 = Q^2 + U^2 + V^2$.
Linearly polarized: $V = 0$; Circularly polarized: $|V| = I$.
Elliptically polarized: $V \neq 0$.

Degree of polarization $= \sqrt{Q^2 + U^2 + V^2}/I$
Degree of linear polarization $= \sqrt{Q^2 + U^2}/I$
Degree of circular polarization $= V/I$

Rayleigh Scattering

Rayleigh scattering is in limit of $x = \frac{2\pi r}{\lambda} \ll 1$.
Particle small compared to wavelength → electric field homogeneous.

Polarizability of matter: E field causes charge separation in particle.
The induced dipole moment $\mathbf{p}$ (charge × distance) is

$$\mathbf{p} = \alpha \mathbf{E}$$

where $\mathbf{E}$ is the applied electric field and α is polarizability.
α depends on particle size and index of refraction (units of volume).

Oscillating E field causes oscillating dipole moment
→ accelerating charges produce electromagnetic radiation.

Radiation produced has same frequency as incident electric field, travels as outgoing spherical wave.
The scattered wave for two polarizations:

$$E_\perp = E_{\perp,0} \frac{e^{-ik(R-ct)}}{R} k^2 \alpha \qquad E_\| = E_{\|,0} \frac{e^{-ik(R-ct)}}{R} k^2 \alpha \cos \Theta$$

where R is distance from particle, $k = \frac{2\pi}{\lambda}$, E_0 is incident field.
k^2 is from acceleration of dipole moment (two time derivatives)
Angular dependence from geometry relative to oscillating dipole moment.

Scattered radiance (square of E field):

$$I_\perp = I_{\perp,0} \frac{(\frac{2\pi}{\lambda})^4 \alpha^2}{R^2} \qquad I_\| = I_{\|,0} \frac{(\frac{2\pi}{\lambda})^4 \alpha^2}{R^2} \cos^2 \Theta$$

Unpolarized incident sunlight $I_{\perp,0} = I_{\|,0} = I_0/2$.
Scattered intensity

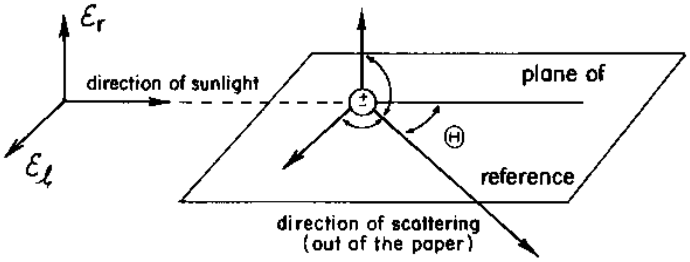
$$I(\Theta) = I_0 \frac{(\frac{2\pi}{\lambda})^4 \alpha^2}{R^2} \frac{1 + \cos^2 \Theta}{2}$$

Rayleigh phase function

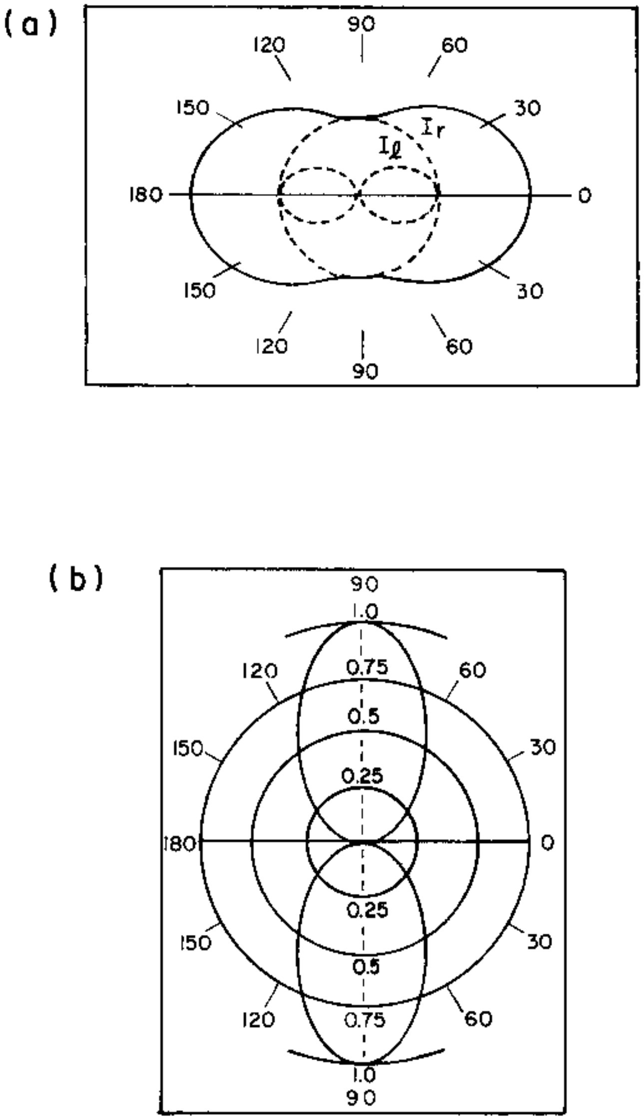
$$P(\Theta) = \frac{3}{4}(1 + \cos^2 \Theta)$$

Rayleigh cross section

$$C_{sca} = \frac{8\pi}{3} \left(\frac{2\pi}{\lambda}\right)^4 |\alpha|^2$$



Geometry for scattering by a dipole. The plane of reference is defined by the incident and scattered directions, with scattering angle Θ between. [Stephens, 1994; Fig. 5.2]



a) Polar diagram of the intensity of Rayleigh scattering for unpolarized incident radiation. b) Polar diagram of the degree of polarization of for Rayleigh scattering. [Stephens, 1994; Fig. 5.3]

Molecular Rayleigh Scattering

Molecular scattering: Rayleigh scattering from air in visible and UV.
For dilute mixtures of gases

$$\alpha = \frac{3}{4\pi N}\frac{m^2-1}{m^2+2} = \frac{m-1}{2\pi N}$$

N is number of molecules per volume.
Index of refraction of air is $m - 1 \approx 6.3 \times 10^{-5} + \text{ small}(\lambda)$.
Since index m is real, molecular scattering has **no absorption**.

Molecular Rayleigh optical depth down to pressure level p in Earth's atmosphere:

$$\tau_{mol} \approx 0.0088 \left(\frac{p}{1013 \text{ mb}}\right) \lambda^{-4.15+0.2\lambda}$$

Complex Index of Refraction

Complex index of refraction, $m = n - i\kappa$, is the material property of a dielectric that determines its radiative properties.
Real part n is bulk refractivity - bends rays (Snells Law).
Imaginary part κ is bulk absorptivity: for bulk material, extinction is $\beta = \frac{4\pi\kappa}{\lambda}$.
Index of refraction depends on material and **wavelength**:
e.g. water in visible $m = 1.33$ (real)
water at $\lambda = 1.55$ cm (10°C) $m = 6.06 - 2.94i$.

Polarizability is complex if index m is complex:
$\rightarrow$ dipole moment is then out of phase with applied field.
Charges oscillate out of phase with E field - ”friction” in material.

Rayleigh Scattering from Spheres

Polarizability of sphere of radius r is

$$\alpha = \frac{m^2-1}{m^2+2} r^3$$

Rayleigh scattering efficiency factors for scattering and absorption:

$$Q_{sca} = \frac{8}{3} x^4 \left| \frac{m^2-1}{m^2+2} \right|^2 \qquad Q_{abs} = -4x \, \mathrm{Im} \left\{ \frac{m^2-1}{m^2+2} \right\}$$

Rayleigh scattering cross sections:

$$C_{sca} = \frac{128\pi^5}{3} \frac{r^6}{\lambda^4} \left| \frac{m^2-1}{m^2+2} \right|^2 \qquad C_{abs} = -\frac{8\pi^2 r^3}{\lambda} \mathrm{Im} \left\{ \frac{m^2-1}{m^2+2} \right\}$$

Scattering $\propto r^6$ (volume^2). Absorption $\propto r^3$ (volume).

Rayleigh Scattering Example

Q_{abs} and Q_{sca} for 0.2 μm radius aerosols at $\lambda = 10\mu$m.
Size parameter $x = \frac{2\pi r}{\lambda} = 2\pi(0.20)/10 = 0.126$.

	$(NH_4)_2SO_4$	NaCl
m	$2.190 - 0.130i$	$1.495 - 5.3 \times 10^{-8} i$
$\frac{m^2-1}{m^2+2}$	$0.56 - 0.037i$	$0.292 - 2.65 \times 10^{-8} i$
Q_{abs}	1.86×10^{-2}	1.33×10^{-8}
Q_{sca}	2.10×10^{-4}	5.66×10^{-5}

$$Q_{sca} = \frac{8}{3}(0.126)^4 \left| 0.56 - 0.037i \right|^2 \qquad Q_{abs} = -4(0.126) \, \mathrm{Im} \left\{ 0.56 - 0.037i \right\}$$

In limit $x \to 0$ **absorption dominates scattering** if $|\mathrm{Im}(m)| > 0$.