

Lecture 5.

Properties of atmospheric aerosols and clouds.

Scattering by small particles: Rayleigh (molecular) scattering.

1. Properties of atmospheric aerosols
2. Properties of clouds and precipitation.
3. Refractive indices of water, ice, and aerosol species.
4. Principles of scattering.
5. Rayleigh scattering.

Required reading:

S: 1.6; 4.1, 4.3; 5.1-5.4, 5.6, 5.7, Petty: 12.1-12.2, 4.1

1. Properties of atmospheric aerosols

Atmospheric aerosols are solid and/or liquid particles suspended in the air with diameters between about 0.002 μm to about 100 μm .

- **Interaction of the particulate matter (aerosols and clouds particles) with electromagnetic radiation is controlled by particle amount, size, composition, mixing state, and shape.**
- Atmospheric particles vary greatly in sources, production mechanisms, sizes, shapes, chemical composition, amount, distribution in space and time, and how long they survive in the atmosphere (i.e., lifetime).

Primary and secondary aerosols:

Primary atmospheric aerosols are particulates that emitted directly into the atmosphere (for instance, sea-salt, mineral aerosols (or dust), volcanic dust, smoke and soot, some organics).

Secondary atmospheric aerosols are particulates that formed in the atmosphere by gas-to-particles conversion processes (for instance, sulfates, nitrates, some organics).

Location in the atmosphere: stratospheric and tropospheric aerosols.

Geographical location: marine, continental, rural, industrial, polar, desert, etc.

Anthropogenic (man-made) and natural aerosols:

Anthropogenic sources: various (biomass burning, gas to particle conversion; industrial processes; agriculture's activities)

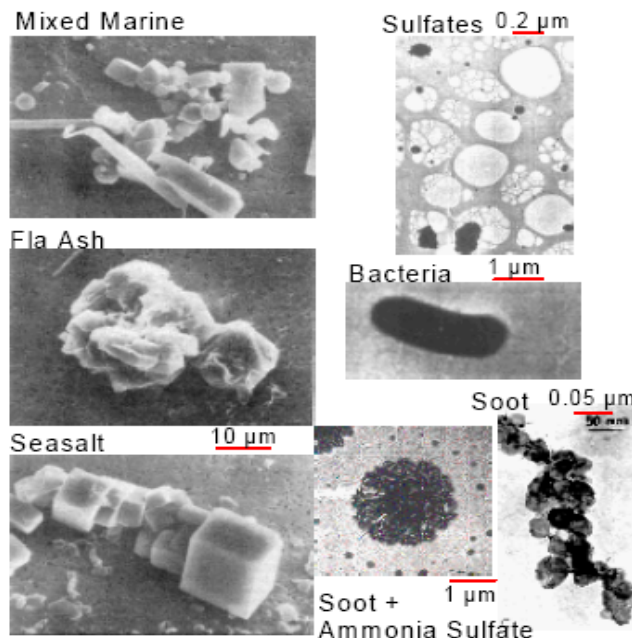
Natural sources: various (sea-salt, dust storm, biomass burning, volcanic debris, gas to particle conversion)

Chemical composition:

Individual chemical species: sulfate (SO_4^{2-}), nitrate (NO_3^-), soot (elemental carbon), sea-salt (NaCl); minerals (e.g., quartz, SiO_2)

Multi-component (MC) aerosols: complex make-up of many chemical species (called internally mixed particles)

Shape:



Spheres: all aqueous aerosol particles (e.g., sulfates, nitrates, etc.)

Complex shapes: dust, soot (i.e., solid particles)

“Classical” representation of particle size spectrum:

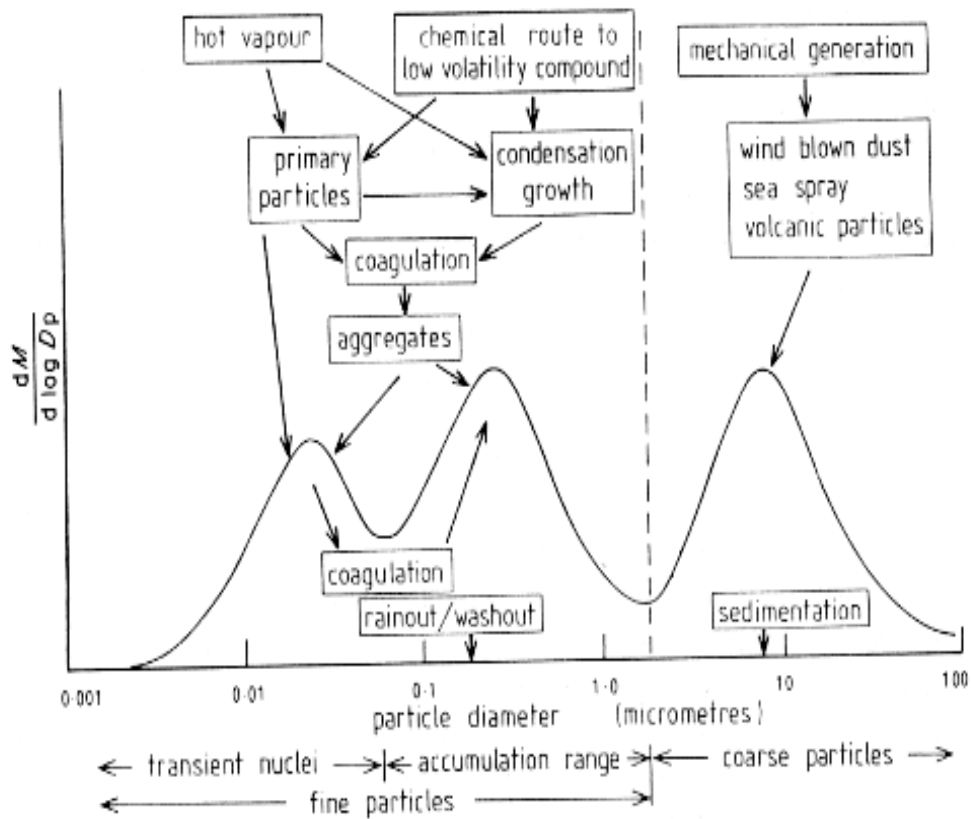


Figure 5.1 Idealized schematic of the distribution of particle surface area of atmospheric aerosol particle (from Whitby and Cantrell, 1976).

NOTE: **fine mode** ($d < 2.5 \mu\text{m}$) and **coarse mode** ($d > 2.5 \mu\text{m}$); fine mode is divided on the **nuclei mode** (about $0.005 \mu\text{m} < d < 0.1 \mu\text{m}$) and **accumulation mode** ($0.1 \mu\text{m} < d < 2.5 \mu\text{m}$).

- **The particle size distribution** of aerosols are often approximated by a sun of three log-normal functions as

$$N(r) = \sum_i \frac{N_i}{\sqrt{2\pi} \ln(\sigma_i) r} \exp\left(-\frac{\ln(r / r_{0,i})^2}{2 \ln(\sigma_i)^2}\right) \quad [5.1]$$

where $N(r)$ is the particle number concentration, N_i is the total particle number concentration of i -th size mode with its median radius $r_{0,i}$ and geometric standard deviation σ_i .

k-moment of a lognormal distribution can be found as

$$\int r^k N(r) dr = N_0 r_0^k \exp(k^2 (\ln \sigma)^2 / 2) \quad [5.2]$$

NOTE: A common approach in the satellite retrieval algorithms is to use a look-up table of aerosol candidate models.

Table 5.1 Aerosol components used in the MISR (Multi-angle Imaging Spectro Radiometer) retrieval algorithm.

Aerosol type	r_0	r_{\min}	r_{\max}	σ	RH (%)	Vary with RH	H_b (km)	H_t (km)	H_s (km)	Shape	ρ g cm ⁻³
Sulfate/nitrate(1) (Accum. Mode)	0.07	0.007	0.7	1.86	0	yes	0	15	2	spheres	1.7
Sulfate/nitrate(2) (Accum. Mode)	0.45	0.05	2.0	1.3	0.3	no	15	30	10	spheres	1.7
Mineral dust (1) (Accum. mode)	0.47	0.05	2.0	1.0	0	no	0	5	2	Prolate/Oblate spheroids	2.6
Mineral dust (2) (Accum. mode)							5	10	10		
Mineral dust (Coarse mode)							0	2	10		
Sea salt (Accum. mode)	0.35	0.05	1.0	2.51	0	yes	0	5	2	spheres	2.2
Sea salt (Coarse mode)	3.3	1.0	20.0	2.03	0	0	2	10			
Black carbon (or soot)	0.012	0.001	0.5	2.0	0	no	0	8	10	spheres	2.3
Carbonaceous	0.13	0.007	2.0	1.8	97	no	0	5	2	spheres	1.8

Here H_b is the base height of the aerosol layer; H_t is the top height of the aerosol layer, and H_s is the scale height of the aerosol layer; ρ is the density of an aerosol particle.

2. Properties of clouds and precipitation

Major characteristics are *cloud type; cloud coverage; liquid water content of cloud; cloud droplet concentration; and cloud droplet size.*

Important properties of clouds:

- Cloud droplet sizes vary from a few micrometers to 100 micrometers with average diameter in 10 to 20 μm range.

- Cloud droplet concentration varies from about 10 cm^{-3} to 1000 cm^{-3} with average droplet concentration of a few hundred cm^{-3} .
- The liquid water content of typical clouds, often abbreviated LWC, varies from approximately 0.05 to $3 \text{ g(water) m}^{-3}$, with most of the observed values in the 0.1 to $0.3 \text{ g(water) m}^{-3}$ region.

NOTE: Clouds cover approximately 60% of the Earth's surface. Average global coverage over the oceans is about 65% and over the land is about 52%.

Table 5.3 Types and properties of clouds.

<i>Type</i>	<i>Height of base (km)</i>	<i>Freq. over oceans (%)</i>	<i>Coverage over oceans (%)</i>	<i>Freq. over land (%)</i>	<i>Coverage over land (%)</i>
Low level:					
Stratocumulus (Sc)	0-2	45	34	27	18
Stratus (St)	0-2	(Sc+St)	(Sc+St)	(Sc+St)	(Sc+St)
Nimbostratus (Ns)	0-4	6	6	6	5
Mid level:					
Altostratus (As)	2-7	46	22	35	21
Altostratus (As)	2-7	(Ac+As)	(Ac+As)	(Ac+As)	(Ac+As)
High level:					
Cirrus (Ci)	7-18	37	13	47	23
Cirrostratus (Cs)	7-18	Ci+Cs+Cc	Ci+Cs+Cc	Ci+Cs+Cc	Ci+Cs+Cc
Cirrocumulus (Cc)	7-18				
Clouds with vertical development					
Cumulus (Cu)	0-3	33	12	14	5
Cumulonimbus (Cb)	0-3	10	6	7	4

- Cloud droplets size distribution is often approximated by a **modified gamma distribution**

$$N(r) = \frac{N_0}{\Gamma(\alpha)r_n} \left(\frac{r}{r_n} \right)^{\alpha-1} \exp(-r/r_n) \quad [5.3]$$

where N_0 is the total number of droplets (cm^{-3}); r_n is the radius that characterizes the distribution ; α is the variance of the distribution, and Γ is the gamma function.

Table 5.4 Characteristics of representative size distributions of some clouds (for $\alpha = 2$)

Cloud type	N_o (cm^{-3})	r_m (μm)	r_{max} (μm)	r_e (μm)	LWC (g m^{-3})
Stratus:					
over ocean	50	10	15	17	0.1-0.5
over land	300-400	6	15	10	0.1-0.5
Fair weather cumulus	300-400	4	15	6.7	0.3
Maritime cumulus	50	15	20	25	0.5
Cumulonimbus	70	20	100	33	2.5
Altostratus	200-400	5	15	8	0.6

- For many practical applications, the optical properties of water clouds are parameterized as a function of the **effective radius** and **liquid water content** (LWC).

The **effective radius** is defined as

$$r_e = \frac{\int \pi r^3 N(r) dr}{\int \pi r^2 N(r) dr} \quad [5.4]$$

where $N(r)$ is the droplet size distribution (e.g., in units $\text{m}^{-3}\mu\text{m}^{-1}$).

NOTE: Mean radius: $r_m = (\alpha + 1) r_n$ Effective radius: $r_e = (\alpha + 3) r_n$

The **liquid water content** (LWC) is defined as

$$LWC = \rho_w V = \frac{4}{3} \rho_w \int \pi r^3 N(r) dr \quad [5.5]$$

➤ **Raindrops**



Nonspherical particles: shape depends of size of a rain drop

- Raindrop size distribution is often represented by the **Marshall-Palmer distribution**:

$$N(r) = N_0 \exp(-2\Lambda r) \quad [5.6]$$

where $N_0 = 8 \times 10^3 \text{ m}^{-3} \text{ mm}^{-1}$, but, in general, N_0 depends on rain type;

Λ is related to the rainfall rate, Rr (in mm/hour) as $\Lambda = 4.1 Rr^{-0.21} \text{ mm}^{-1}$

➤ **Ice crystals**

- Depending on the atmospheric conditions, clouds may consist of ice crystals (e.g., cirrus clouds) or ice crystal/water droplet mixtures.

Ice crystals often have the hexagonal structure with sizes on the order of several hundred micrometers. However, a large variety of shapes (called habits) and sizes of ice crystals have been reported: Plates - nearly flat hexagon; Columns - elongated, flat bottoms; Needles - elongated, pointed bottoms; Dendrites - elongated arms (six), snowflake shape.

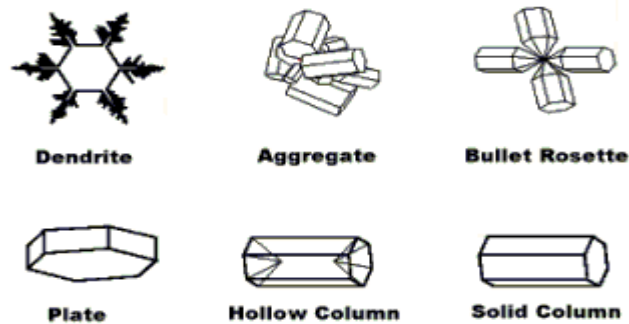


Figure 5.2 Examples of ice habits: shapes depend on temperature and relative humidity.

3. Refractive indices of water, ice, and aerosol species.

Refractive index (or optical constants), $m = n - ik$, is the material properties of dielectric that determines its radiative properties. In general, each material has its own spectral refractive index. The imaginary part k of the refractive index determines the absorption of the wave as it propagates through the medium; the real part n of the refractive index gives the phase velocity of propagation.

- It is believed that the refractive indices of the medium (bulk material) apply down to the smallest atmospheric aerosol particles.

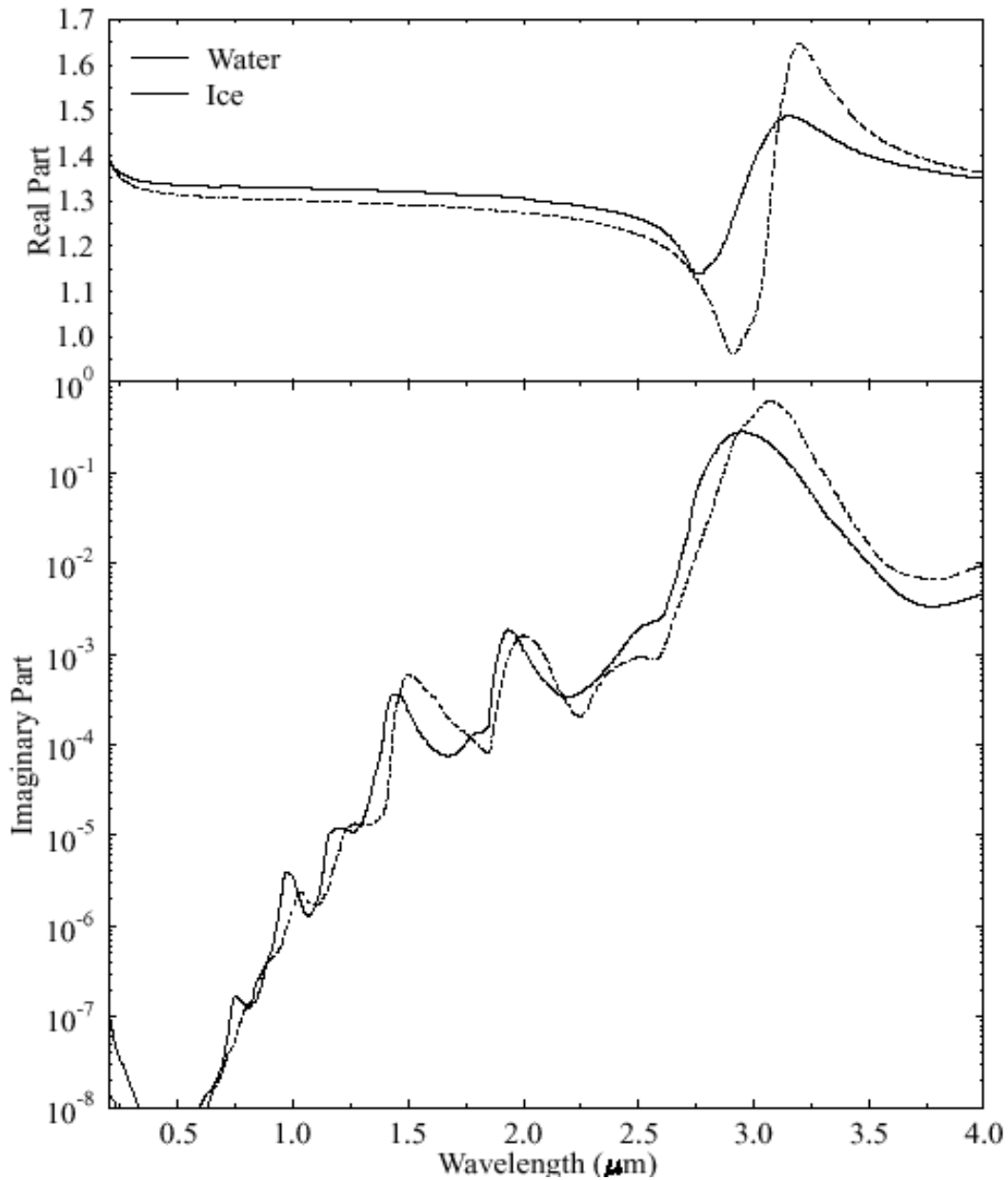


Figure 5.3 The refractive index of water and ice in the visible and near-IR.

NOTE: water has the low imaginary part in the visible => negligible absorption by water drops in the visible

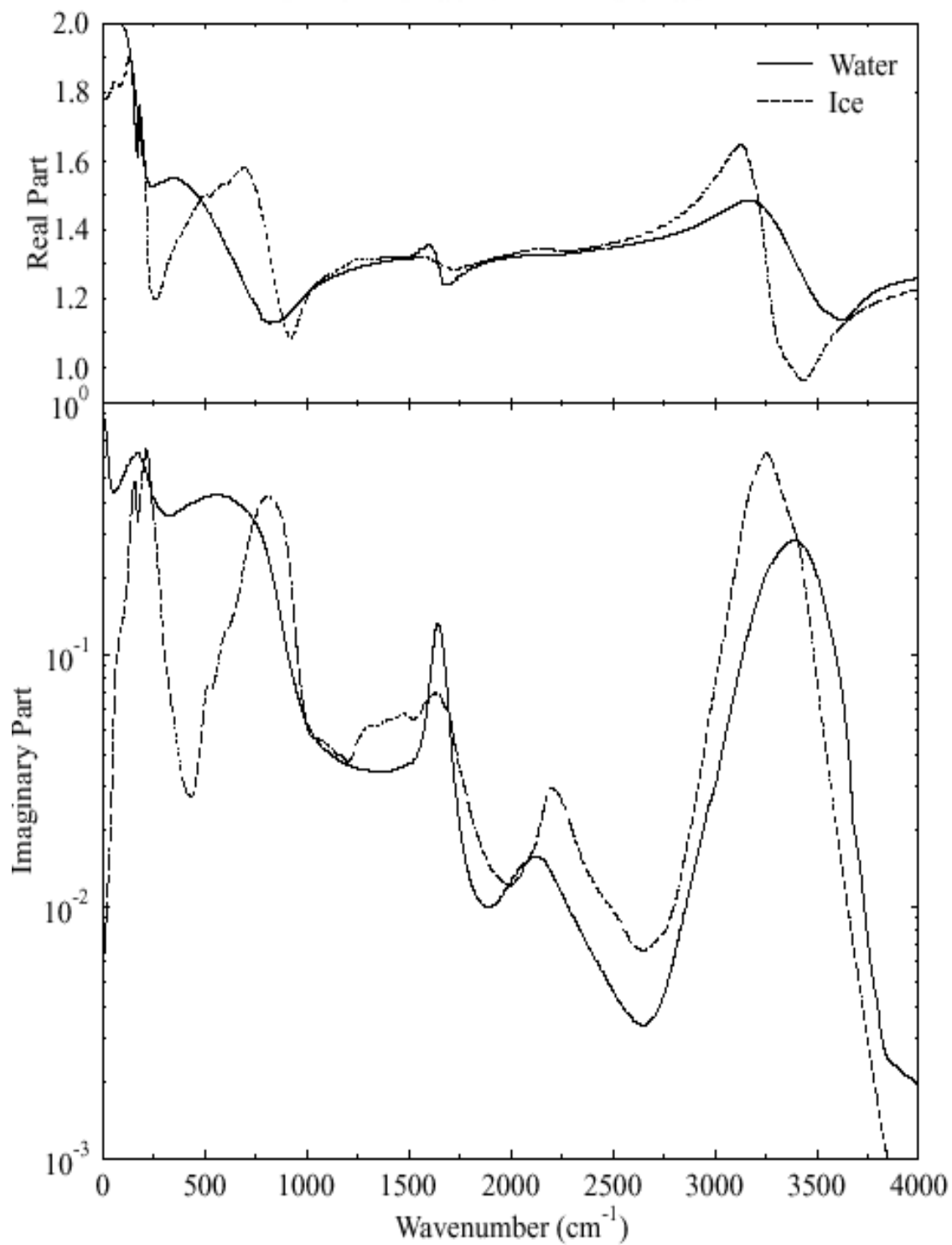


Figure 5.4 The refractive index of water and ice in the IR.

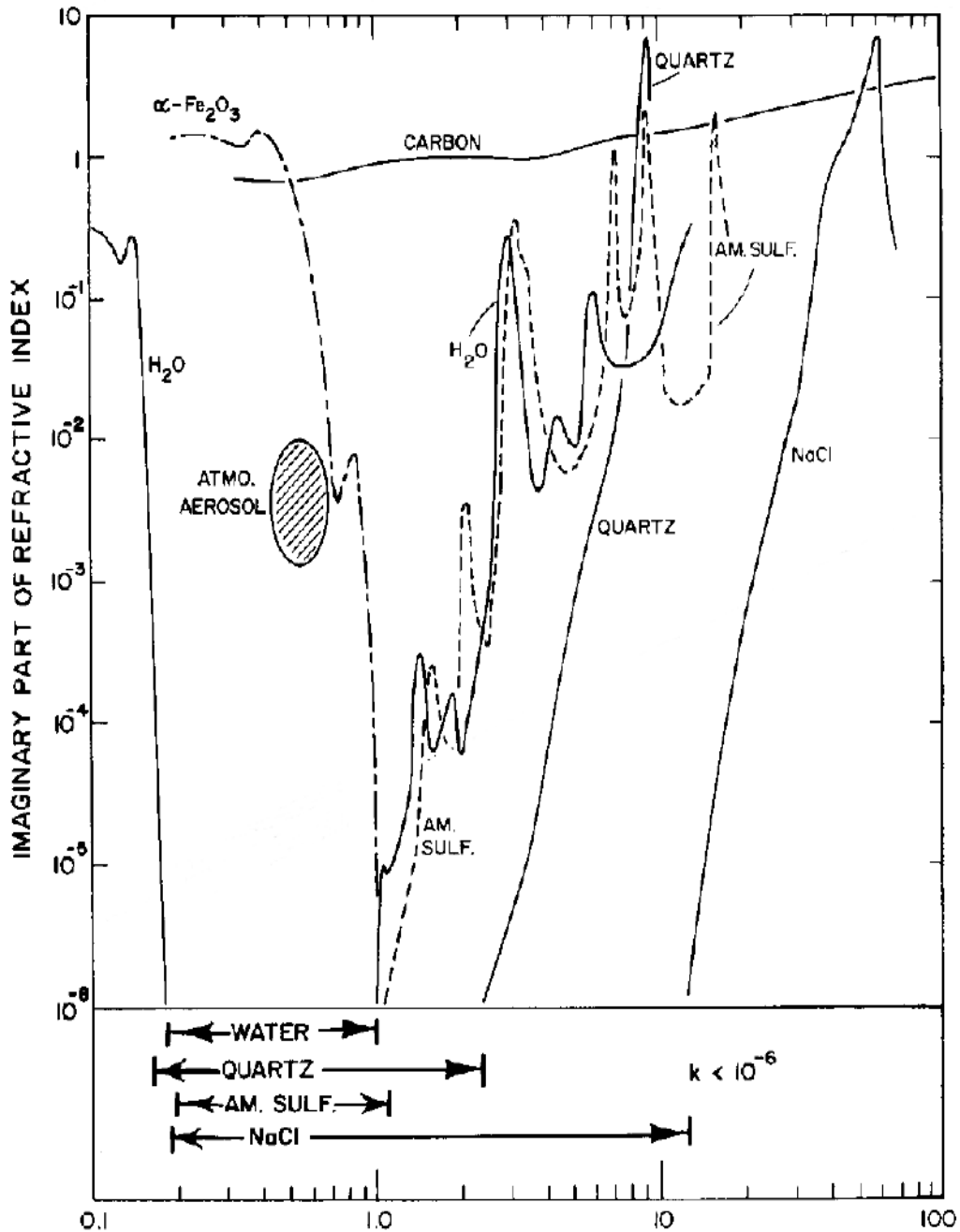


Figure 5.5 “A “classical plot” showing the imaginary part of the refractive indexes of some aerosol materials (Bohren and Huffman, Fig.5.16).

NOTE: Main absorbing species in the solar spectrum are black carbon (soot) and hematite (dust), but in the thermal IR various species have high imaginary parts of the refractive index.

- Aerosol particles often consist of several chemical species (called the **internal mixture**).

There are several approaches (called **mixing rules**) to calculate the **effective refractive index** m_e of the internally mixed particles using the refractive indices of the individual species:

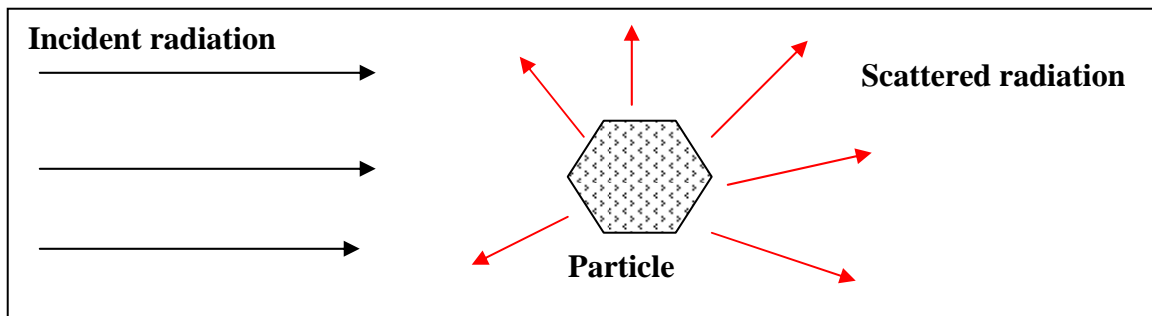
Volume weighted mixing:

$$m_e = \sum_j m_j f_j \quad [5.7]$$

where m_j is the refractive index of j-species and f_j is its volume fraction.

NOTE: Other commonly used mixing rules are the Bruggeman and Maxwell-Garnett.

4. Principles of scattering.



Consider a single arbitrary particle consisted of many individual dipoles. The incident electromagnetic field induces dipole oscillations. The dipoles oscillate at the frequency of the incident field and therefore **scatter radiation** in all directions. In a given direction of observation, the total scattered field is a superposition of the scattered wavelets of these dipoles, accounting for their phase difference: scattering by the dipoles is coherent (i.e., there is a definite relation between phases).

Scattering phase function $P(\cos\Theta)$ is defined as a non-dimensional parameter to describe the angular distribution of the scattered radiation as

$$\frac{1}{4\pi} \int_{\Omega} P(\cos \Theta) d\Omega = 1 \quad [5.8]$$

where Θ is called the **scattering angle** between the direction of incidence and observation.

NOTE: Another form of [5.8]

$$\frac{1}{4\pi} \int_0^{2\pi} \int_0^\pi P(\cos\Theta) \sin\Theta d\Theta d\varphi = 1 \quad [5.9]$$

NOTE: The phase function is often expressed as

$$P(\cos\Theta) = P(\theta', \varphi', \theta, \varphi')$$

where (θ', φ') and (θ, φ) are the spherical coordinates of incident beam and direction of observation. Using the spherical geometry it can be shown (see S: Appendix 1) that

$$\cos(\Theta) = \cos(\theta')\cos(\theta) + \sin(\theta')\sin(\theta) \cos(\varphi' - \varphi)$$

The **asymmetry factor** g is defined as

$$g = \frac{1}{2} \int_{-1}^1 P(\cos\Theta) \cos\Theta d(\cos\Theta) \quad [5.10]$$

$g=0$ for isotropic scattering

Forward scattering refers to the observations directions for which $\Theta < \pi/2$:

$g > 0$ scattering in the forward direction

Backward scattering refers to the observations directions for which $\Theta > \pi/2$:

$g < 0$ scattering in the backward direction

Scattering domains:

Rayleigh scattering: $2\pi r/\lambda \ll 1$, and the refractive index m is arbitrary (applies to scattering by molecules and small aerosol particles)

Rayleigh-Gans scattering: $(m - 1) \ll 1$ (not useful for atmospheric application)

Mie-Debye scattering: $2\pi r/\lambda$ and m are both arbitrary but for spheres only (applies to scattering by aerosol and cloud particles)

Geometrical optics: $2\pi r/\lambda \gg 1$ and m is real (applies to scattering by large cloud droplets and ice crystals).

5. Rayleigh scattering.

- Because the sizes of atmospheric molecules are much smaller than the wavelengths of solar and IR radiation, scattering by atmospheric gases is the Rayleigh scattering domain.
- In the Rayleigh scattering approximation, a molecule (or a small particle) is considered as an individual dipole.

Consider a small homogeneous spherical particle (e.g., a molecule) with size smaller than the wavelength of incident radiation \vec{E}_0 . Let \vec{p}_0 be the induced dipole moment, then from the classical electromagnetic theory we have

$$\vec{p}_0 = \alpha \vec{E}_0 \quad [5.11]$$

where α is the **polarizability** of the particle.

NOTE: Do not confuse the polarization of the medium with polarization associated with the EM wave.

The scattered electric field at the large distance r (called far field scattering) from the dipole is given (in cgs units) by

$$\vec{E} = \frac{1}{c^2} \frac{1}{r} \frac{\partial \vec{p}}{\partial t} \sin(\gamma) \quad [5.12]$$

where γ is the angle between the scattered dipole moment \vec{p} and the direction of observation. In oscillating periodic field, the dipole moment is given in terms of induced dipole moment by

$$\vec{p} = \vec{p}_0 \exp(-ik(r - ct)) \quad [5.13]$$

and thus the electrical field is

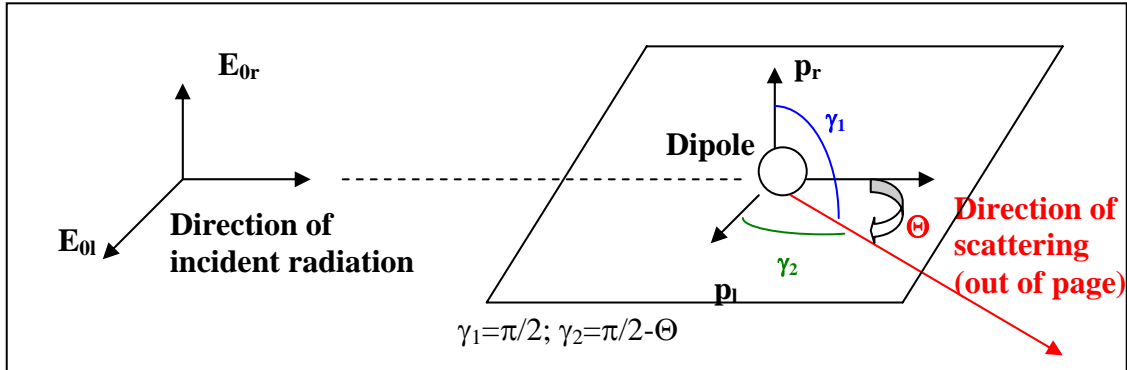
$$\vec{E} = -\vec{E}_0 \frac{\exp(-ik(r - ct))}{r} k^2 \alpha \sin(\gamma) \quad [5.14]$$

Decomposing the electrical vector on two orthogonal components perpendicular and parallel to **the plane of scattering** (a plane containing the incident and scattering beams),

We have

$$E_r = -E_{0r} \frac{\exp(-ik(r-ct))}{r} k^2 \alpha \sin(\gamma_1) \quad [5.15]$$

$$E_l = -E_{0l} \frac{\exp(-ik(r-ct))}{r} k^2 \alpha \sin(\gamma_2) \quad [5.16]$$



Using that

$$I = \frac{1}{\Delta\Omega} \frac{c}{4\pi} |E|^2 \quad [5.17]$$

the perpendicular and parallel intensities (or linear polarized intensities) are

$$I_r = I_{0r} k^4 \alpha^2 / r^2 \quad [5.18]$$

$$I_l = I_{0l} k^4 \alpha^2 \cos^2(\Theta) / r^2 \quad [5.19]$$

Using that the natural light (incident beam) is not polarized ($I_{0r} = I_{0l} = I_0/2$) and that $k = 2\pi/\lambda$, we have

$$I = I_r + I_l = \frac{I_0}{r^2} \alpha^2 \left(\frac{2\pi}{\lambda} \right)^4 \frac{1 + \cos^2(\Theta)}{2} \quad [5.20]$$

Eq.[5.20] gives the intensity scattered by molecules (Rayleigh scattering) for unpolarized incident light.

Rayleigh scattering phase function **for incident unpolarized radiation is**

$$P(\cos(\Theta)) = \frac{3}{4}(1 + \cos^2(\Theta)) \quad [5.21]$$

Eq.[5.20] may be rewritten in the form

$$I(\cos(\Theta)) = \frac{I_0}{r^2} \alpha^2 \frac{128 \pi^5}{3 \lambda^4} \frac{P(\Theta)}{4 \pi} \quad [5.22]$$

Eq. [5.22] may be rewritten in the terms of **the scattering cross section**

$$I(\cos(\Theta)) = \frac{I_0}{r^2} \sigma_s \frac{P(\Theta)}{4 \pi} \quad [5.23]$$

Here the scattering cross section (in units of area) by a single molecule is

$$\sigma_s = \alpha^2 \frac{128 \pi^5}{3 \lambda^4} \quad [5.24]$$

The **polarizability** is given by the Lorentz-Lorenz formula

$$\alpha = \frac{3}{4 \pi N_s} \left(\frac{m^2 - 1}{m^2 + 2} \right) \quad [5.25]$$

where N_s is the number of molecules per unit volume and $m=n - ik$ in the refractive index.

For air molecules in solar spectrum: n is about 1 but depends on λ , and $k=0$.

Thus the polarizability can be approximated as

$$\alpha \approx \frac{1}{4 \pi N_s} (n^2 - 1) \quad [5.26]$$

Therefore, the scattering cross section of an air molecule becomes

$$\sigma_s = \frac{8 \pi^3 (n^2 - 1)^2}{3 \lambda^4 N_s^2} f(\delta) \quad [5.27]$$

where $f(\delta)$ is the correction factor for the anisotropic properties of air molecules, defined as $f(\delta) = (6+3\delta)/(6-7\delta)$ and $\delta=0.035$

Using this scattering cross section, one can calculate the **optical depth** of the entire atmosphere **due to molecular scattering** as

$$\tau(\lambda) = \sigma_s(\lambda) \int_0^{\text{top}} N(z) dz \quad [5.28]$$

NOTE: The Rayleigh scattering cross section (Eq.[5.24]) and hence optical depth are inversely proportional to the fourth power of the wavelength (=> blue color of the sky)

Approximation of molecular Rayleigh optical depth (i.e., optical depth due to molecular scattering) down to pressure level p in the Earth's atmosphere:

$$\tau(\lambda) \approx 0.0088 \left(\frac{p}{1013 \text{ mb}} \right) \lambda^{-4.15+0.2\lambda} \quad [5.29]$$

- Rayleigh scattering results in the sky polarization. The degree of linear polarization is

$$LP(\Theta) = -\frac{Q}{I} = -\frac{I_l - I_r}{I_l + I_r} = -\frac{\cos^2 \Theta - 1}{\cos^2 \Theta + 1} = \frac{\sin^2 \Theta}{\cos^2 \Theta + 1} \quad [5.30]$$