

## Lecture 5.

### Molecular (Rayleigh) scattering.

#### Scattering and absorption by aerosol and cloud particles: Mie theory.

1. Beer-Bouguer-Lambert law (Extinction law).
2. Basics of scattering.
3. Molecular (Rayleigh) scattering.
4. Properties of aerosol and clouds.
5. Scattering and absorption by aerosol and cloud particles.
6. Remote sensing applications based on measurements of direct solar radiation (see also Lab 3).

#### Required reading:

S: 1.6; 4.1, 4.3; 5.1-5.4, 5.6, 5.7, Appendix 1;

Petty: 4, 7, 12

#### Additional reading

S: 4.2; 5.5

#### Advanced reading

Bohren, C. F., and D. R. Huffman, *Absorption and scattering of light by small particles*. John Wiley&Sons, New York, pp. 531, 1983.

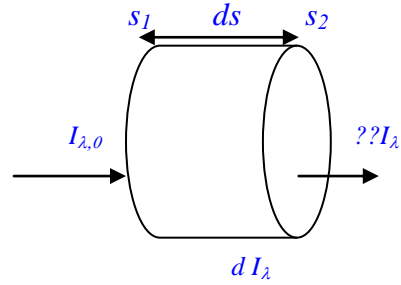
### 1. Beer-Bouguer-Lambert law (Extinction law) .

The fundamental law of extinction is the **Beer-Bouguer-Lambert law**, which states that the extinction process is linear in the intensity of radiation and amount of matter, provided that the physical state (i.e., T, P, composition) is held constant.

Consider a small volume  $\Delta V$  of infinitesimal length  $ds$  and unit area  $\Delta A$  containing optically active matter (gases, aerosols, and/or cloud drops). Thus, the change of intensity along a path  $ds$  is proportional to the amount of matter in the path:

For extinction 
$$dI_\lambda = -k_{e,\lambda} I_\lambda ds \quad [5.1a]$$

For emission: 
$$dI_\lambda = k_{a,\lambda} B_\lambda ds \quad [5.1b]$$



where  $\kappa_{e,\lambda}$  is the **volume extinction coefficient** ( $\text{LENGTH}^{-1}$ ), where  $\kappa_{a,\lambda}$  is the **volume absorption coefficient** ( $\text{LENGTH}^{-1}$ ), and  $B_\lambda$  is the **Planck function**. Here the amount is the particle number concentration.

Recall that extinction is due to scattering and absorption:

$$k_{e,\lambda} = k_{s,\lambda} + k_{a,\lambda} \quad [5.2]$$

Integrating Eq.[5.1a], we have

$$I_\lambda = I_{0,\lambda} \exp\left(-\int_{s_1}^{s_2} k_{e,\lambda}(s) ds\right) = I_{0,\lambda} \exp(-\tau_\lambda) \quad [5.3]$$

where  $I_{0,\lambda}$  and  $I_\lambda$  are the incident and transmitted intensities, respectively.

**NOTE:** optical depth  $\tau$  is unitless.

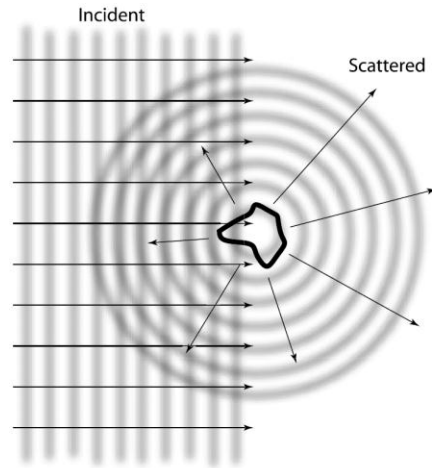
**NOTE:** “same name”: **optical depth = optical thickness = optical path**

**Transmission function** is defined as

$$T_\lambda = I_\lambda / I_{0,\lambda} = \exp(-\tau_\lambda) \quad [5.4]$$

**UNITS:** transmission function is unitless (between 0 and 1)

## 2. Basics of scattering.



**Figure 5.1** Simplified visualization of scattering of an incident EM wave by a particle.

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 certain 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

$$\boxed{\frac{1}{4\pi} \int_{\Omega} P(\cos \Theta) d\Omega = 1} \quad [5.5]$$

where  $\Theta$  is called the **scattering angle** between the direction of incidence and direction of observation.

Another form of Eq.[5.5]

$$\frac{1}{4\pi} \int_0^{2\pi} \int_0^{\pi} P(\cos \Theta) \sin \Theta d\Theta d\varphi = 1 \quad [5.6]$$

**NOTE:** The phase function is often written with the notations  $(\theta', \varphi', \theta, \varphi)$  or  $(\mu', \varphi', \mu, \varphi)$

$$P(\cos \Theta) = P(\theta', \varphi', \theta, \varphi) \quad [5.7]$$

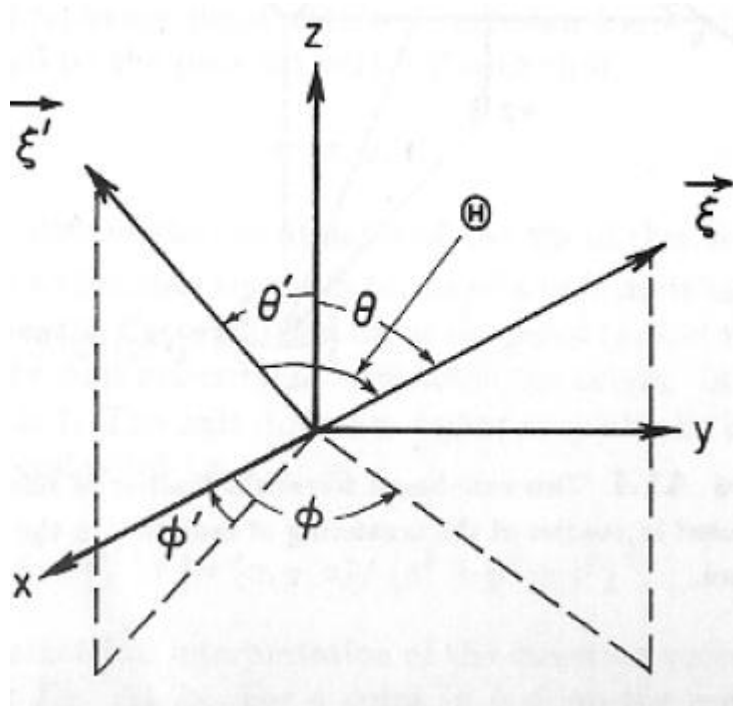
$$P(\cos \Theta) = P(\mu', \varphi', \mu, \varphi)$$

where  $(\theta', \varphi')$  and  $(\theta, \varphi)$  are the spherical coordinates of incident beam and direction of observation:  $\theta'$  and  $\theta$  are the zenith angles, and  $\varphi'$  and  $\varphi$  are the azimuth angles; and  $\mu = \cos(\theta)$  and  $\mu' = \cos(\theta')$ .

The scattering angle  $\Theta$  is expressed in terms of  $(\theta', \varphi', \theta, \varphi)$  as (see S: Appendix 1)

$$\cos(\Theta) = \cos(\theta')\cos(\theta) + \sin(\theta')\sin(\theta) \cos(\varphi' - \varphi) \text{ or}$$

$$\cos(\Theta) = \mu' \mu + (1 - \mu'^2)^{1/2} (1 - \mu^2)^{1/2} \cos(\varphi' - \varphi)$$



**Figure A1.2** from Stephens, Appendix 1, p.468

The **asymmetry factor  $g$**  is defined as

$$g = \frac{1}{2} \int_{-1}^1 P(\cos \Theta) \cos \Theta d(\cos \Theta) \quad [5.8]$$

$g=0$  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 applications)

**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, rain, drops and ice crystals).

**3. Molecular (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, point 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

$$\vec{p}_0 = \alpha \vec{E}_0 \quad [5.9]$$

where  $\alpha$  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

$$\vec{E} = \frac{1}{c^2} \frac{1}{r} \frac{\partial \vec{p}}{\partial t} \sin(\gamma) \quad [5.10]$$

where  $\gamma$  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

$$\vec{p} = \vec{p}_0 \exp(-ik(r - ct)) \quad [5.11]$$

and thus the electrical field is

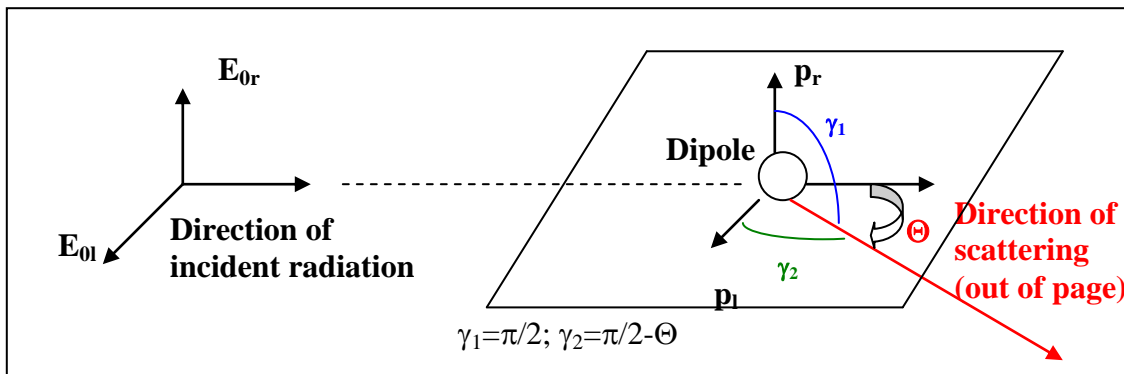
$$\vec{E} = -\vec{E}_0 \frac{\exp(-ik(r - ct))}{r} k^2 \alpha \sin(\gamma) \quad [5.12]$$

(Note here  $k$  denotes  $2\pi/\lambda$ ).

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

$$E_r = -E_{0r} \frac{\exp(-ik(r - ct))}{r} k^2 \alpha \sin(\gamma_1) \quad [5.13]$$

$$E_l = -E_{0l} \frac{\exp(-ik(r - ct))}{r} k^2 \alpha \sin(\gamma_2) \quad [5.14]$$



Using that

$$I = \frac{1}{\Delta\Omega} \frac{c}{4\pi} |E|^2 \quad [5.15]$$

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

$$I_r = I_{0r} k^4 \alpha^2 / r^2 \quad [5.16]$$

$$I_l = I_{0l} k^4 \alpha^2 \cos^2(\Theta) / r^2 \quad [5.17]$$

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

$$I = I_r + I_l = \frac{I_0}{r^2} \alpha^2 \left( \frac{2\pi}{\lambda} \right)^4 \frac{1 + \cos^2(\Theta)}{2} \quad [5.18]$$

**Eq. [ 5.18] gives the intensity scattered by molecules (Rayleigh scattering) for unpolarized incident light.**

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

$$P(\cos(\Theta)) = \frac{3}{4} (1 + \cos^2(\Theta)) \quad [5.19]$$

Eq. [ 5.18] may be rewritten in the form

$$I(\cos(\Theta)) = \frac{I_0}{r^2} \alpha^2 \frac{128\pi^5}{3\lambda^4} \frac{P(\Theta)}{4\pi} \quad [5.20]$$

Eq. [5.20] may be rewritten in the terms of **the scattering cross section**

$$I(\cos(\Theta)) = \frac{I_0}{r^2} \sigma_s \frac{P(\Theta)}{4\pi} \quad [5.21]$$

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

$$\sigma_s = \alpha^2 \frac{128\pi^5}{3\lambda^4} \quad [5.22]$$

The **polarizability** of a point dipole is given by the Lorentz-Lorenz formula

$$\alpha = \frac{3}{4\pi N_s} \left( \frac{m^2 - 1}{m^2 + 2} \right) \quad [5.23]$$

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

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

Thus the polarizability can be approximated as

$$\alpha \approx \frac{1}{4\pi N_s} (n^2 - 1) \quad [5.24]$$

Therefore, the scattering cross section of an air molecule becomes

$$\sigma_s = \frac{8\pi^3 (n^2 - 1)^2}{3\lambda^4 N_s^2} f(\delta) \quad [5.25]$$

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 estimate the **optical depth** of the entire atmosphere **due to molecular scattering** as

$$\tau(\lambda) = \sigma_s(\lambda) \int_0^{top} N(z) dz \quad [5.26]$$

**NOTE:** The Rayleigh scattering cross section (Eq.[5.22]) and hence optical depth are inversely proportional to the fourth power of the wavelength ( $\Rightarrow$  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:

$$\tau(\lambda) \approx 0.0088 \left( \frac{P}{1013mb} \right) \lambda^{-4.15+0.2\lambda} \quad [5.27]$$

where the wavelength  $\lambda$  is in  $\mu m$ .



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

$$LP(\Theta) = -\frac{Q}{I} = -\frac{I_l - I_r}{I_l + I_r} = -\frac{\cos^2 \Theta - 1}{\cos^2 \Theta + 1} = \frac{\sin^2 \Theta}{\cos^2 \Theta + 1} \quad [5.28]$$

#### 4. Properties of aerosol and clouds.

**Interaction of particulates (aerosols, cloud drops, ice crystals, rain drops, etc.) with electromagnetic radiation is controlled by the particle amount, size, composition (refractive index), and shape.**

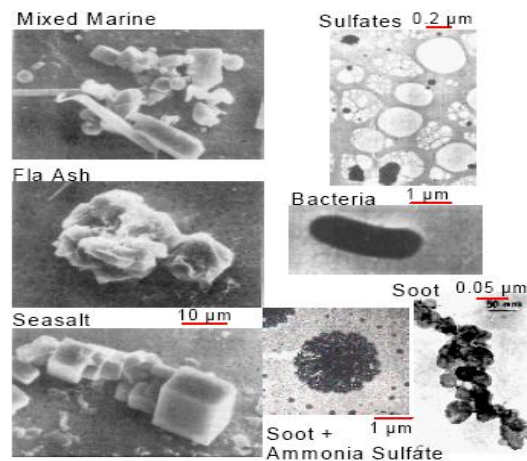
- **Atmospheric aerosols** are solid and/or liquid (or mixed phase) particles suspended in the air with diameters between about 0.002 μm to about 50-100 μm.

#### Chemical composition:

**Individual chemical species:** sulfate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), soot (elemental carbon or black carbon), sea-salt (NaCl); minerals (e.g., quartz, SiO<sub>4</sub>, clays, feldspar, etc.)

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

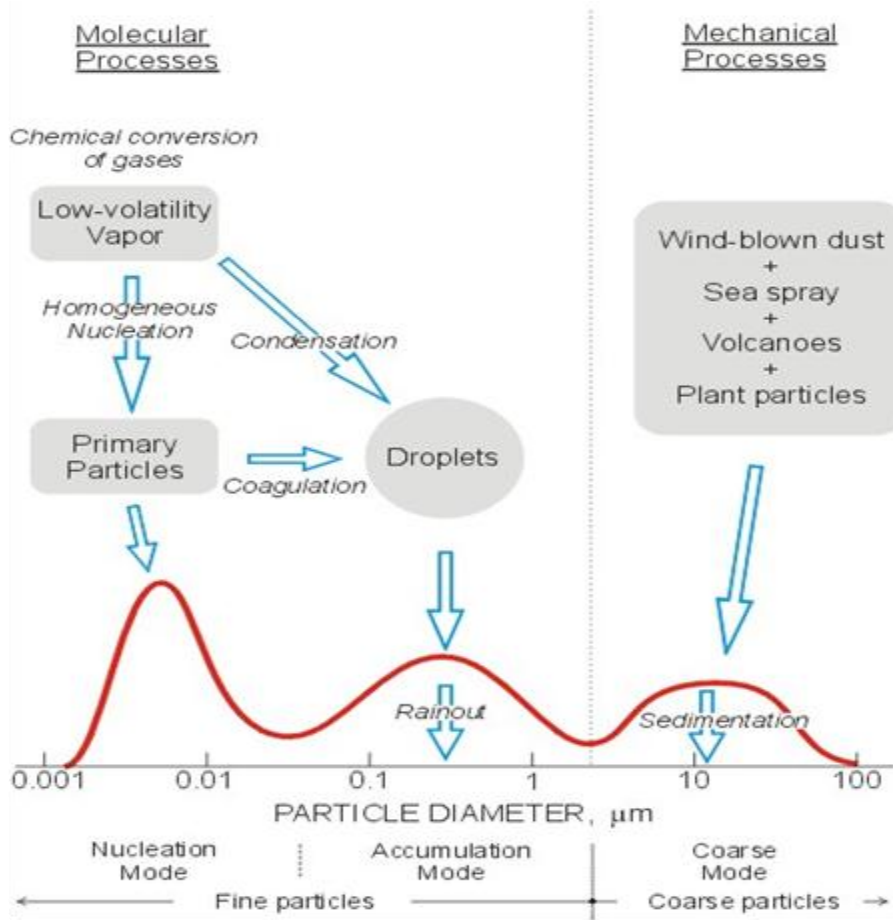
#### Shape:



**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 5.2** Idealized schematic of the size distribution 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 sum of three log-normal functions as

$$N(r) = \sum_i \frac{N_i}{\sqrt{2\pi} \ln(\sigma_i)} \frac{1}{r} \exp\left(-\frac{\ln(r/r_{0,i})^2}{2 \ln(\sigma_i)^2}\right) \quad [5.29]$$

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  $\sigma_i$ .

Property of the log-normal function: the k-moment is

$$\int r^k N(r) dr = N_0 r_0^k \exp(k^2 (\ln \sigma)^2 / 2) \quad [5.30]$$

**NOTE:** Eq [5.30] helps to quickly calculate the mass, volume, and surface area size distribution from the particle number size distribution.

➤ **Clouds:**

- ✓ Cloud droplet sizes vary from a few micrometers to 100 micrometers with average diameter in the 10 to 20  $\mu\text{m}$  range.
- ✓ Cloud droplet concentration varies from about  $10 \text{ cm}^{-3}$  to  $1000 \text{ cm}^{-3}$  with an average droplet concentration of a few hundred  $\text{cm}^{-3}$ .
- ✓ 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.

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

$$N(r) = \frac{N_0}{\Gamma(\alpha) r_n} \left( \frac{r}{r_n} \right)^{\alpha-1} \exp(-r / r_n) \quad [5.31]$$

where  $N_0$  is the total number of droplets ( $\text{cm}^{-3}$ );  $r_n$  in the radius that characterizes the distribution ;  $\alpha$  in the variance of the distribution, and  $\Gamma$  is the gamma function.

**Table 5.1** Characteristics of representative size distributions of some clouds (for  $\alpha = 2$ )

Cloud type	$N_0$ ( $\text{cm}^{-3}$ )	$r_m$ ( $\mu\text{m}$ )	$r_{\max}$ ( $\mu\text{m}$ )	$r_e$ ( $\mu\text{m}$ )	LWC ( $\text{g m}^{-3}$ )
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

**NOTE:** 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

$$r_e = \frac{\int \pi r^3 N(r) dr}{\int \pi r^2 N(r) dr} \quad [5.32]$$

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

$$LWC = \rho_w V = \frac{4}{3} \rho_w \int \pi r^3 N(r) dr \quad [5.33]$$

➤ **Raindrops**



Nonspherical particles: shape depends on size of a rain drop.

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

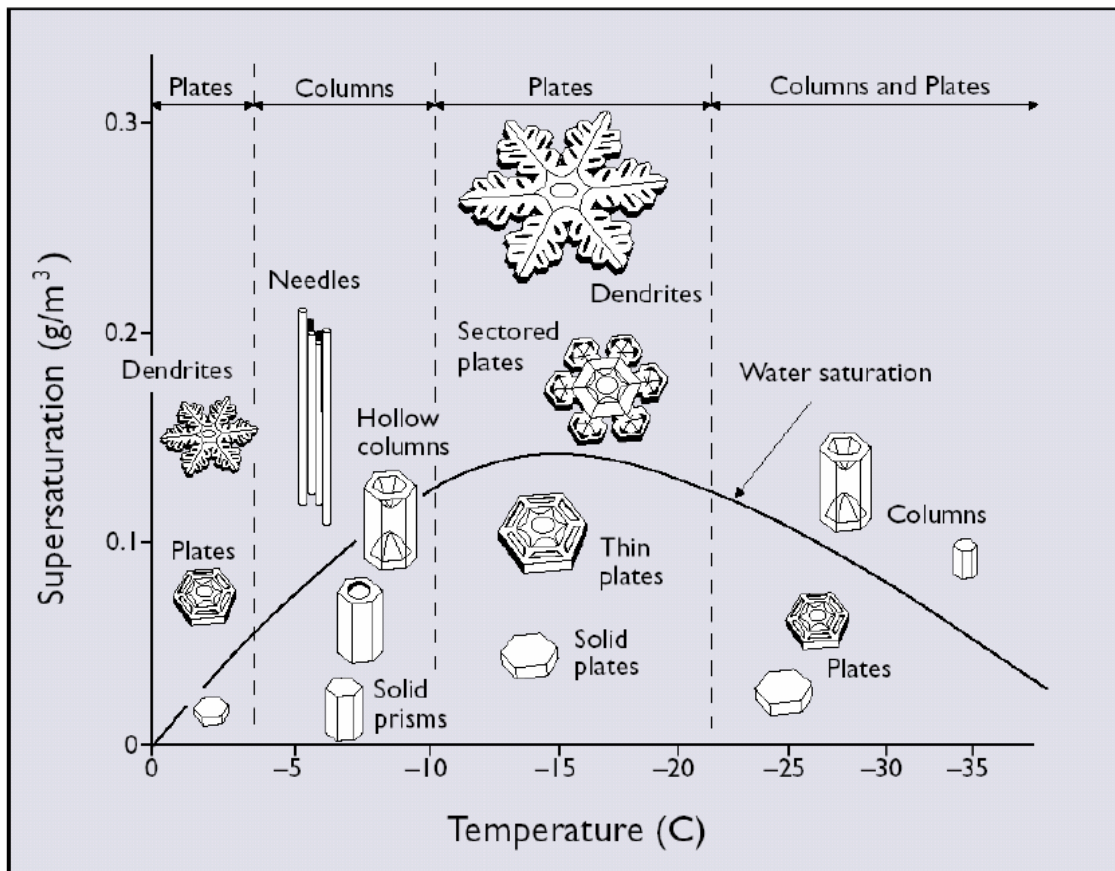
$$N(r) = N_0 \exp(-2\Lambda r) \quad [5.34]$$

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

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

➤ **Ice crystals:**

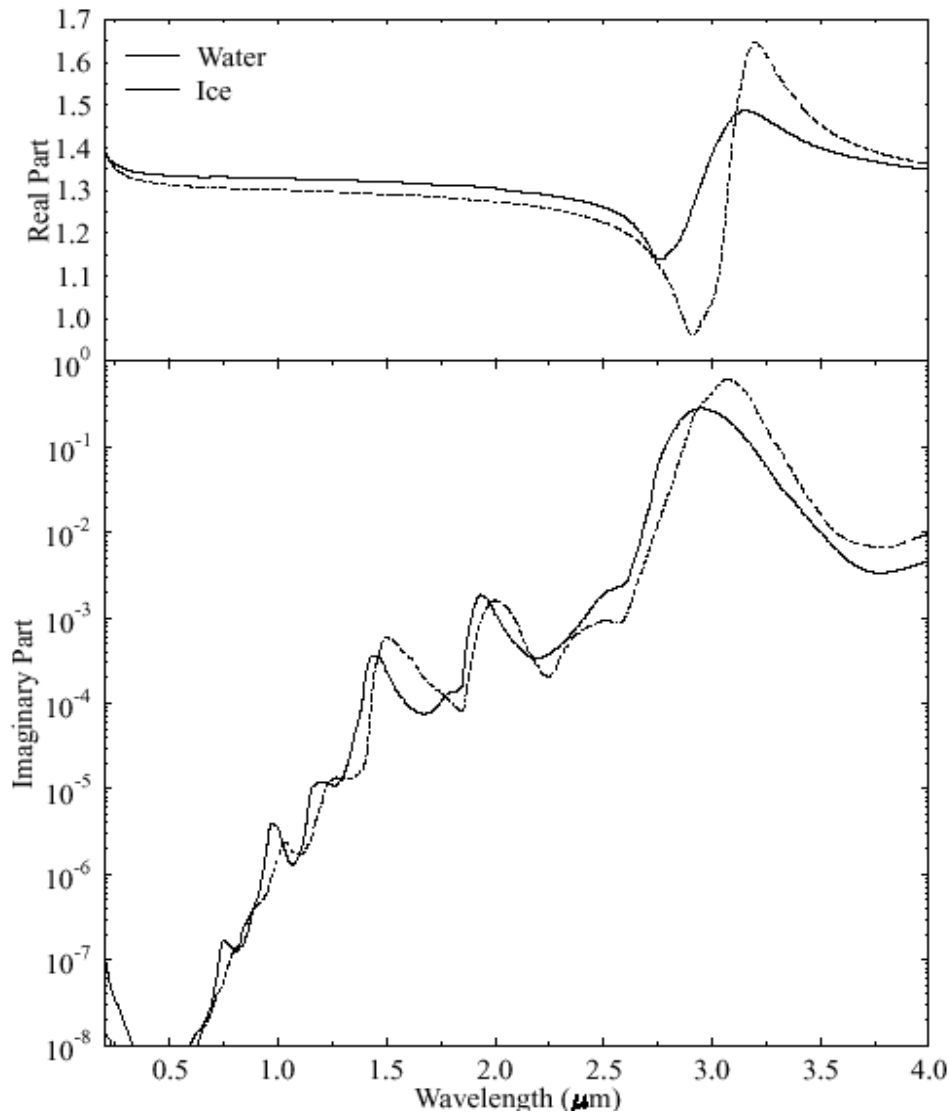
- ✓ Sizes range from ~5-10  $\mu\text{m}$  to a few mm
- ✓ Exhibit a large variety of shapes (called habits): for instance, plates - nearly flat hexagon; columns - elongated, flat bottoms; needles - elongated, pointed bottoms; dendrites - elongated arms (six), snowflake shape.



**Figure 5.3** Ice crystal's shape as a function of supersaturation and temperature – a “classical” view

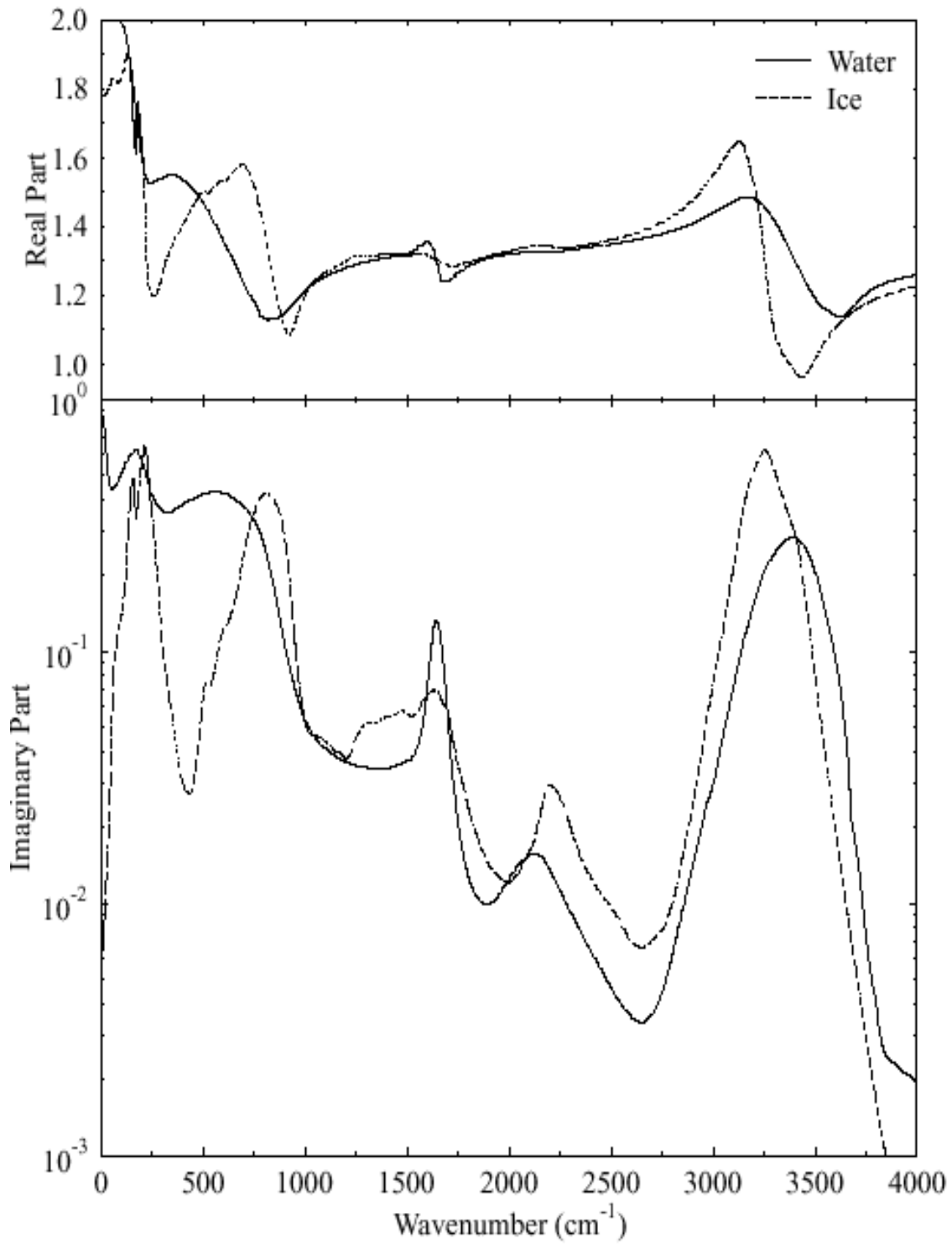
➤ Refractive indices of water, ice, and aerosol species.

**Refractive index (or optical constants)**,  $m=n - i\kappa$ , is the material properties of dielectric that determines its radiative properties. In general, each material has its own spectral refractive index. The imaginary part  $\kappa$  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.

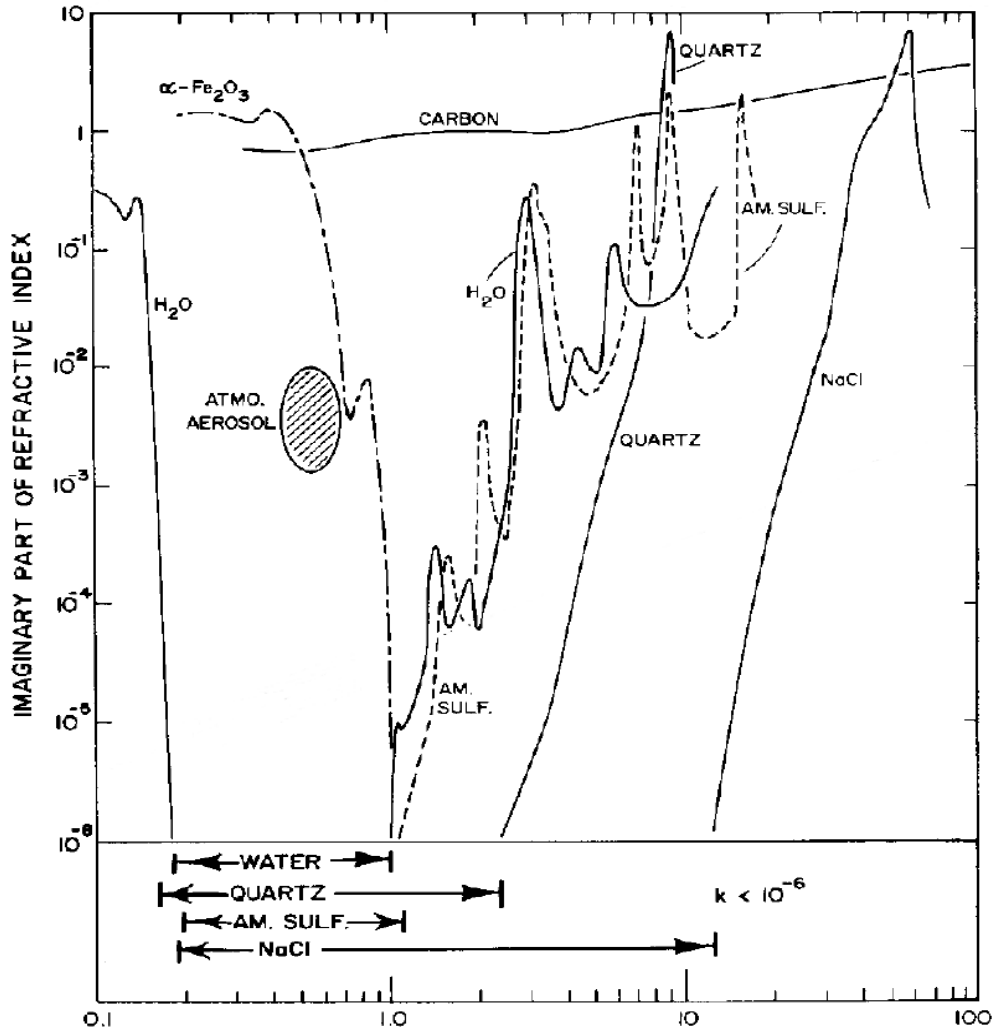


**Figure 5.4** The refractive index of water and ice in the visible and near-IR.

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



**Figure 5.5** The refractive index of water and ice in the IR.



**Figure 5.6** “Classical plot” of the imaginary part of the refractive indexes of some aerosol materials as a function of wavelength (in  $\mu\text{m}$ ) (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.

**NOTE:** Aerosol particles often consist of several chemical species (called the **internal mixture**). There are several approaches (called **mixing rules**) to calculate an **effective refractive index**  $m_e$  of the internally mixed particles by using the refractive indices of the individual species. For instance, **Volume weighted mixing**:

$$m_e = \sum_j m_j f_j \quad [5.35]$$

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



## 5. Scattering and absorption by aerosol and cloud particles: Mie theory.

**Mie theory** describes the scattering and absorption of electromagnetic radiation by spherical particles through solving the Maxwell equations.

**NOTE:** Mie theory is also called **Lorenz-Mie theory** or **Lorenz-Mie-Debye theory**.

===== **Mie theory outline** =====

### General outline of Mie theory:

#### *Key Assumptions:*

- i) Particle is a **sphere**;
- ii) Particle is **homogeneous** (therefore it is characterized by a **single refractive index**  $m=n - i\kappa$  at a given wavelength);

**NOTE:** Mie theory requires the relative refractive index that is the refractive index of a particle divided by the refractive index of a medium. For air  $m$  is about 1, so one needs to know the refractive index of the particle (i.e., refractive index of the material of which the particle is composed).

**NOTE:** If a particle has complex chemical composition such as some atmospheric aerosols, the effective refractive index must be calculated at a given wavelength.

- Mie theory calculates the scattered electromagnetic field at all points within the particle (called internal field) and at all points of the homogeneous medium in which the particle is embedded. For all practical applications in the atmosphere, light scattering observations are carried out in the **far-field zone** (i.e., at the large distances from a particle).

In the far-field zone (i.e., at the large distances  $R$  from a sphere), the solution of the wave equation can be obtained as

$$\begin{bmatrix} E_l^s \\ E_r^s \end{bmatrix} = \frac{\exp(-ikR + ikz)}{ikR} \begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} \begin{bmatrix} E_l^i \\ E_r^i \end{bmatrix} \quad [5.36]$$

here  $k = 2\pi/\lambda$ ,  $E_l^i$  and  $E_r^i$  are the parallel and perpendicular components of incident electrical field, and  $E_l^s$  and  $E_r^s$  are the parallel and perpendicular components of scattered electrical field,

$\begin{bmatrix} S_2(\Theta) & S_3(\Theta) \\ S_4(\Theta) & S_1(\Theta) \end{bmatrix}$  is the **amplitude scattering matrix** (unitless)

**For spheres:**  $S_3(\Theta) = S_4(\Theta) = 0$ , and thus Eq.[6.1] gives

$$\begin{bmatrix} E_l^s \\ E_r^s \end{bmatrix} = \frac{\exp(-ikR + ikz)}{ikR} \begin{bmatrix} S_2(\Theta) & 0 \\ 0 & S_1(\Theta) \end{bmatrix} \begin{bmatrix} E_l^i \\ E_r^i \end{bmatrix} \quad [5.37]$$

**Eq.[5.37] is a fundamental equation of scattered radiation by a sphere including polarization.**

**Mie theory solution for the scattering amplitudes:**

$$S_1(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n \pi_n(\cos \Theta) + b_n \tau_n(\cos \Theta)] \quad [5.38]$$

$$S_2(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [b_n \pi_n(\cos \Theta) + a_n \tau_n(\cos \Theta)] \quad [5.39]$$

where  $\pi_n$  and  $\tau_n$  are **Mie angular functions**

$$\pi_n(\cos \Theta) = \frac{1}{\sin(\Theta)} P_n^1(\cos \Theta) \quad [5.40]$$

$$\tau_n(\cos \Theta) = \frac{d}{d\Theta} P_n^1(\cos \Theta) \quad [5.41]$$

where  $P_n^1$  are the associated Legendre polynomials.

Mie theory also gives the **scattering phase matrix**  $P(\Theta)$  that relates the Stokes parameters  $\{I_0, Q_0, U_0 \text{ and } V_0\}$  of incident radiation field and the Stokes parameters  $\{I, Q, U \text{ and } V\}$  of scattered radiation:

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi r^2} P \begin{bmatrix} I_o \\ Q_o \\ U_o \\ V_o \end{bmatrix} \quad [5.42]$$

where

$$P = \begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{22} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{34} \\ 0 & 0 & P_{34} & P_{44} \end{bmatrix} \quad [5.43]$$

**NOTE:** In general, for a particle of any shape, the scattering phase matrix consists of 16 independent elements, but for a sphere this number reduces to four.

**For spheres:**  $P_{22} = P_{11}$  and  $P_{44} = P_{33}$

Thus **for spheres**, Eq.[5.42] reduces to

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi r^2} \begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{11} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{34} \\ 0 & 0 & P_{34} & P_{33} \end{bmatrix} \begin{bmatrix} I_o \\ Q_o \\ U_o \\ V_o \end{bmatrix} \quad [5.44]$$

and each element of the scattering phase matrix is expressed via the scattering amplitudes  $S_1(\Theta)$  and  $S_2(\Theta)$ .

$P_{11}(\Theta) = P(\Theta)$  is the scattering phase function of a particle.

From Mie theory it follows that the **extinction cross-section** of a particle is

$$\sigma_e = \frac{4\pi}{k^2} \text{Re}[S(0^0)] \quad [5.45]$$

But for the forward direction (i.e.  $\Theta = 0^0$ ) from Eqs.[4.38]-[5.39], we have

$$S_1(0^0) = S_2(0^0) = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1)(a_n + b_n)$$

-----**END of Mie theory outline**-----

- **Efficiencies (or efficiency factors)** for extinction, scattering and absorption are defined as

$$Q_e = \frac{\sigma_e}{\pi r^2} \quad Q_s = \frac{\sigma_s}{\pi r^2} \quad Q_a = \frac{\sigma_a}{\pi r^2} \quad [5.46]$$

where  $\pi r^2$  is the area of a article with radius  $r$ .

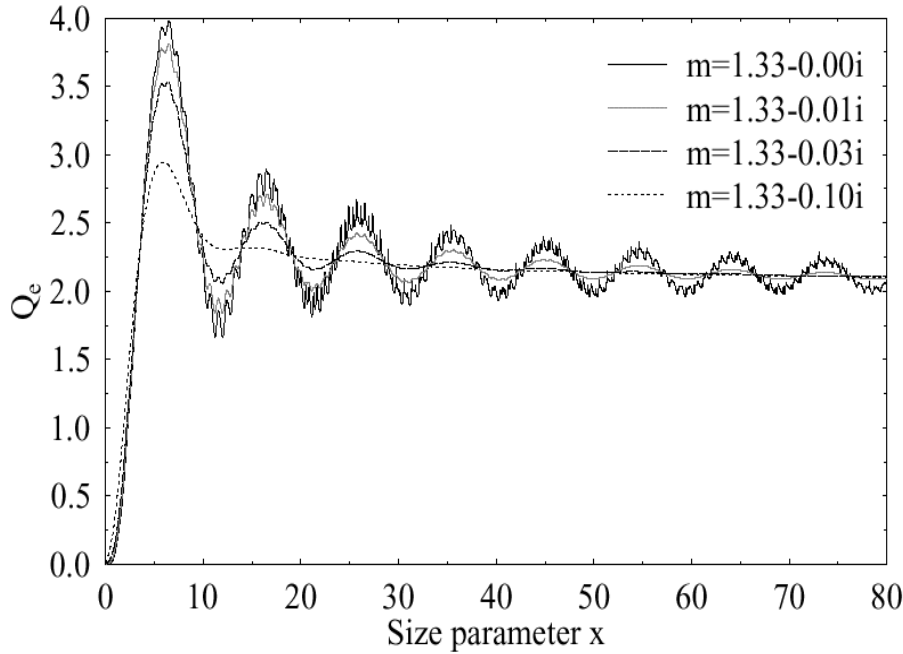
Mie theory gives the solution for  $Q_e$ ,  $Q_s$  and  $Q_a$  in terms coefficient  $a_n$  and  $b_n$  (i.e., coefficients in the expressions for the scattering amplitudes  $S_1(0^0)$  and  $S_2(0^0)$ ).

$$Q_e = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \text{Re}[a_n + b_n] \quad [5.47]$$

$$Q_s = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) [|a_n|^2 + |b_n|^2] \quad [5.48]$$

and the absorption efficiency can be calculated as

$$Q_a = Q_e - Q_s \quad [5.49]$$



**Figure 5.7** Examples of  $Q_e$  calculated with the Mie theory for several refractive indexes.

**Some highlights of Mie theory results:**

- Extinction efficiency vs. size parameter  $x$  (assuming NO ABSORPTION):
  - 1) small in Rayleigh limit:  $Q_e \propto x^4$
  - 2) largest  $Q_e$  when particles and wavelength have similar size
  - 3)  $Q_e \rightarrow 2$  in the geometric limit ( $x \rightarrow \infty$ )
  - 4) Oscillations (see Fig.5.7) from interference of transmitted and diffracted waves
- Period in  $x$  of interference oscillations depends on the refractive index. Absorption reduces interference oscillations and kills ripple structure.
- Scattering and absorption efficiencies vs. size parameter with ABSORPTION:
 

As  $x \rightarrow \infty$ :  $Q_s \rightarrow 1$  and , entering rays are absorbed inside particle. Smaller imaginary part of the refractive index requires larger particle to fully absorb internal rays.
- Scattering phase function: forward peak height increases dramatically with  $x$ . For single particles – number of oscillations in  $P(\Theta)$  increases with  $x$ .

**For a single spherical particle, Mie theory gives the extinction, scattering and absorption cross-sections (and efficiency factors), the scattering amplitudes and phase matrix.**

***Integration over the particle size distribution:***

If particles have the size distribution  $N(r)$ , the volume extinction, scattering and absorption coefficients (in units  $\text{LENGTH}^{-1}$ ) are determined as

$$k_e = \int_{r_1}^{r_2} \sigma_e(r)N(r)dr = \int_{r_1}^{r_2} \pi r^2 Q_e N(r)dr \quad [5.50]$$

$$k_s = \int_{r_1}^{r_2} \sigma_s(r)N(r)dr = \int_{r_1}^{r_2} \pi r^2 Q_s N(r)dr \quad [5.51]$$

$$k_a = \int_{r_1}^{r_2} \sigma_a(r)N(r)dr = \int_{r_1}^{r_2} \pi r^2 Q_a N(r)dr \quad [5.52]$$

Similar to Eq.[4.12], the optical depth of an aerosol layer (between  $s_1$  and  $s_2$ ) is

$$\tau_{\lambda}(s_1, s_2) = \int_{s_1}^{s_2} k_{e,\lambda} ds = \int_{s_1}^{s_2} M k_{m,e,\lambda} ds$$

where  $k_{m,e,\lambda}$  is the mass extinction coefficient and M is the mass of particles.

**NOTE:** Mass coefficients = volume coefficients/particle mass concentration, M

The **single scattering albedo** gives the percentage of light which will be scattered in a single scattered event and it is defined as

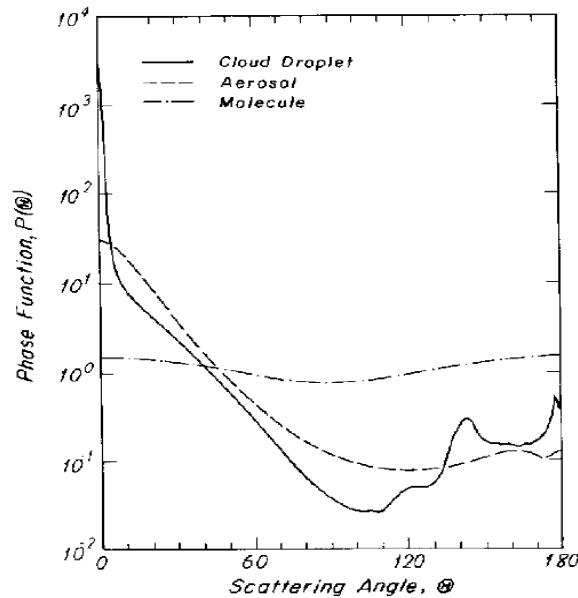
$$\omega_0 = \frac{k_s}{k_e} \quad [5.53]$$

**NOTE:** No absorption (conservative scattering):  $\omega_0 = 1$

No scattering:  $\omega_0 = 0$

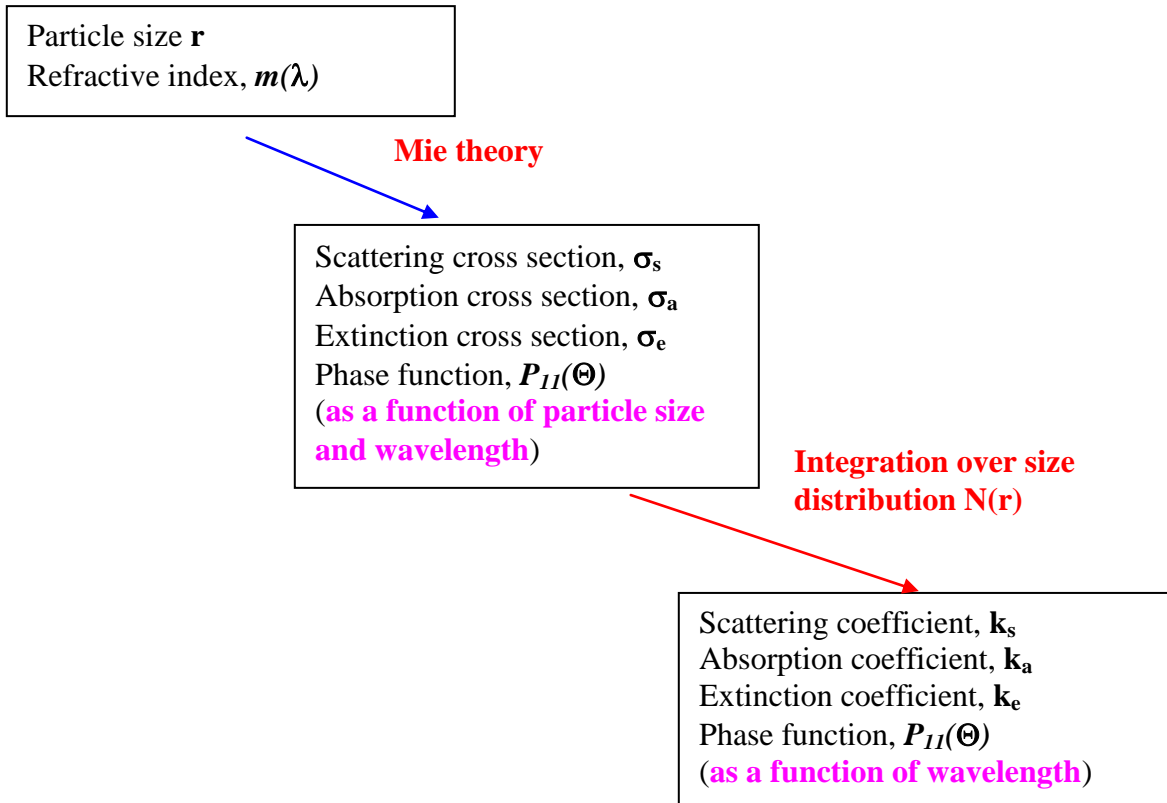
**Scattering phase function** of particles with the size distribution N(r):

$$P(\Theta) = \frac{\int_{r_1}^{r_2} P(\Theta, r) \sigma_s N(r) dr}{\int_{r_1}^{r_2} \sigma_s N(r) dr} \quad [5.54]$$



**Figure 5.8** Examples of representative scattering phase functions (at a wavelength of 0.5  $\mu\text{m}$ ) for aerosol and clouds particles. The molecular (Rayleigh) scattering phase function is also shown for comparison.

**How to calculate optical characteristics of an ensemble of spherical particles:**



- **Optical properties of the external mixture** (i.e., the mixture of several types of particles)

$$k_e = \sum_i k_e^i \qquad k_s = \sum_i k_s^i \qquad k_a = \sum_i k_a^i \qquad [5.55]$$

where  $k_e^i$ ,  $k_s^i$  and  $k_a^i$  are calculated for each particle type characterized by its particle size distribution  $N_i(r)$  and a refractive index (or effective refractive index)  $m_i$ .

**NOTE:** Do not sum the single scattering albedo and scattering phase functions!!! See below Eq.[5.57-5.58] how to do it.

➤ **How to calculate the effective optical properties of an atmospheric layer consisting of gas and aerosols (and clouds):**

In general, an atmospheric layer consists of molecules, aerosols and/or cloud particles.

Thus, one needs to calculate the **effective optical properties** of the layer:

**Effective (also called total) optical depth:**

$$\tau_{\lambda} = \tau_{a,\lambda}^M + \tau_{s,\lambda}^M + \tau_{a,\lambda}^A + \tau_{s,\lambda}^A \quad [5.56]$$

where  $\tau_{a,\lambda}^M$  and  $\tau_{s,\lambda}^M$  are optical depth due to absorption by gases and molecular (Rayleigh) scattering, respectively;

$\tau_{a,\lambda}^A$  and  $\tau_{s,\lambda}^A$  are optical depth due to absorption and scattering by aerosol particles, respectively.

**Effective single scattering albedo:**

$$\omega_{0,\lambda} = \frac{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A}{\tau_{\lambda}} \quad [5.57]$$

**Effective scattering phase function:**

$$P_{\lambda}(\Theta) = \frac{\tau_{s,\lambda}^M P_{\lambda}^M(\Theta) + \tau_{s,\lambda}^A P_{\lambda}^A(\Theta)}{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A} \quad [5.58]$$

**Effective asymmetry parameter:**

$$g_{\lambda} = \frac{\tau_{s,\lambda}^A g_{\lambda}^A}{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A} \quad [5.59]$$

## **6. Remote sensing applications based on measurements of direct solar radiation**

**Direct radiation** is a part of the radiation field that has survived the extinction passing a layer with optical depth  $\tau$  and it obeys the Beer-Bouguer-Lambert law (or Extinction law):

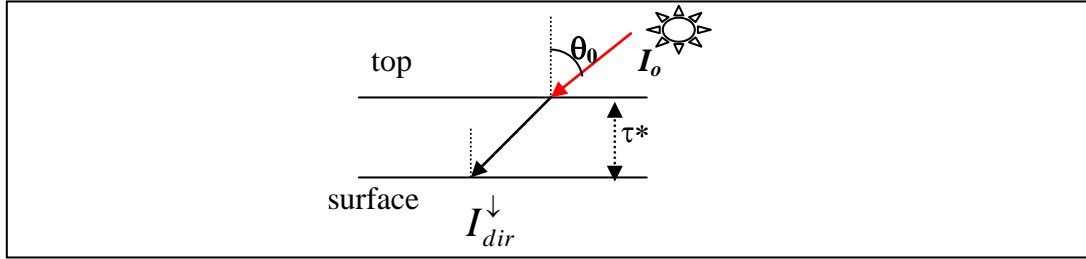
$$I_{dir}^{\downarrow} = I_0 \exp(-\tau / \mu_0) \quad [5.60]$$

where  $I_0$  is the incident intensity at a given wavelength at the top of a layer and  $\mu_0$  is a cosine of the incident zenith angle  $\theta_0$  ( $\mu_0 = \cos(\theta_0)$ ).



**Applying the Extinction law to direct solar radiation:**

$I_0$  is the solar intensity at a given wavelengths at the top of the atmosphere



Thus direct solar radiation reaching the surface is

$$I_{dir}^{\downarrow} = I_0 \exp(-\tau / \mu_0) \quad [5.61]$$

where  $\tau$  is the **optical depth on the entire atmosphere**.

**NOTE:** Optical depth is defined along vertical coordinate (i.e., altitude  $z$ ) in the atmosphere

**Retrieval of aerosol optical depth from ground-based sunphotometer measurement:**

A sunphotometer (a narrow-field of view radiometer that tracks the sun) measures the direct solar radiation attenuated by the atmosphere:

$$F_{dir,\lambda}^{\downarrow} = F_{0,\lambda} \exp(-\tau_{\lambda}^* / \mu_0) \quad [5.62]$$

where  $F_{dir,\lambda}^{\downarrow}$  is the downward direct solar flux reaching the surface (i.e., the downward direct solar intensity integrated over the viewing angle of the sunphotometer) and  $\tau_{\lambda}^*$  is the optical depth of the entire atmospheric column, i.e.

$$\tau_{\lambda}^* = \tau_{\lambda}(z_{top}, 0) = \int_0^{z_{top}} k_{e,\lambda}(z) dz \quad [5.63]$$

For cloud-free atmospheric conditions,  $\tau_{\lambda}^*$  is due to attenuation by aerosols, molecular (Rayleigh) scattering and gaseous absorption (e.g.,  $O_3$  and  $NO_2$  depending on  $\lambda$ ). Thus

$$\tau_{\lambda}^* = \tau_{a,\lambda}^M + \tau_{s,\lambda}^M + \tau_{\lambda}^A \quad [5.64]$$

From Eq.[5.62] we have

$$\ln(F_{dir,\lambda}^\downarrow) = \ln(F_{0,\lambda}) - \tau_\lambda^* / \mu_0 \quad [5.65]$$

and

$$\tau_\lambda^* = \mu_0 \{ \ln(F_{0,\lambda}) - \ln(F_{dir,\lambda}^\downarrow) \} \quad [5.66]$$

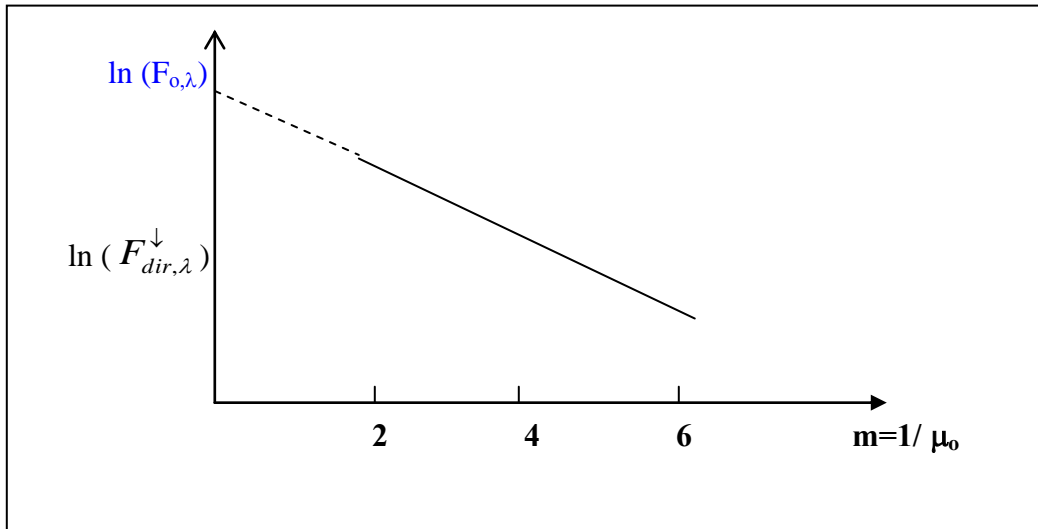
Thus

$$\tau_\lambda^A = \mu_0 \{ \ln(F_{0,\lambda}) - \ln(F_{dir,\lambda}^\downarrow) \} - \{ \tau_{a,\lambda}^M + \tau_{s,\lambda}^M \} \quad [5.67]$$

**NOTE:** To retrieve the aerosol optical depth, one needs to correct for Rayleigh scattering and gaseous absorption.

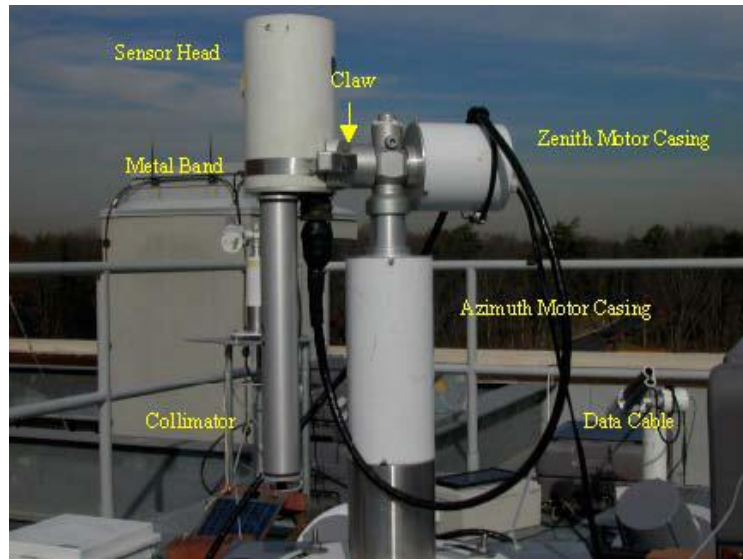
**Other applications:**

i) If the aerosol optical depth remains constant during the day, Eq.[5.67] enables to measure the solar flux  $F_{0,\lambda}$  by plotting measured  $F_{dir,\lambda}^\downarrow$  vs.  $\mu_0$  (called the Langley plot).



ii) If the aerosol optical depth is known or negligibly small, Eq.[5.67] gives the optical depth due to absorption of gases (used in the retrievals of O<sub>3</sub> and H<sub>2</sub>O column amount).

**Examples:** AERONET - <http://aeronet.gsfc.nasa.gov/> - (AErosol RObotic NETwork) program is a federation of ground-based remote sensing aerosol networks established originally by NASA and CNRS and expanded by many international agencies, universities, etc. The program provides a long-term, continuous and readily accessible public data of aerosol optical properties (and water vapor column amount) for research and validation of satellite aerosol optical depth retrievals.



**Figure 5.9** Cimel Sunphotometer operated by AERONET. Typical spectral bands (channels) of the Cimel sunphotometer (central wavelengths): 340 nm, 380 nm, 440nm, 500 nm, 675 nm, 870 nm and 1020nm.

Measurements of aerosol spectral optical depth can be used to compute the **Angstrom parameter** (also called the **Angstrom exponent**) which relates to the aerosol particle size distribution: for measurements of optical depth  $\tau_{\lambda_1}$  and  $\tau_{\lambda_2}$  taken at two different wavelengths  $\lambda_1$  and  $\lambda_2$  respectively, the Angstrom exponent  $\alpha$  is given by

$$\alpha = - \frac{\ln \frac{\tau_{\lambda_1}}{\tau_{\lambda_2}}}{\ln \frac{\lambda_1}{\lambda_2}}$$

or

$$\frac{\tau_{\lambda_1}}{\tau_{\lambda_2}} = \left( \frac{\lambda_1}{\lambda_2} \right)^{-\alpha} \quad [5.68]$$

**NOTE:** For air (pure molecular scattering):  $\alpha = 4$  and for large particles (compared to the wavelength)  $\alpha = 0$