

Lecture 6.

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

Main radiation law (Beer-Bouguer- Lambert law)

1. Scattering and absorption by aerosol and cloud particles.
2. Main radiation law (Beer-Bouguer-Lambert law).
3. Remote sensing applications based on measurements of the direct solar radiation.

Required reading:

S: 5.3-5.6; Petty: 12, 7.2, 7.4.2

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. 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**.

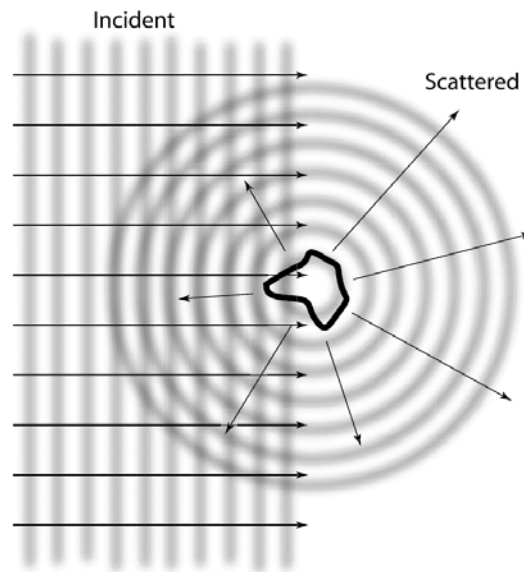


Figure 6.1 Simplified visualization of scattering of an incident EM wave by a particle.

===== 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** $m=n - ik$ 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

$$\begin{bmatrix} E_t^s \\ E_r^s \end{bmatrix} = \frac{\exp(-ikR + ikz)}{ikR} \begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} \begin{bmatrix} E_t^i \\ E_r^i \end{bmatrix} \quad [6.1]$$

here $k = 2\pi/\lambda$, E_t^i and E_r^i are the parallel and perpendicular components of incident electrical field, and E_t^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: $S_3(\Theta) = S_4(\Theta) = \mathbf{0}$, and thus Eq.[6.1] gives

$$\begin{bmatrix} E_t^s \\ E_r^s \end{bmatrix} = \frac{\exp(-ikR + ikz)}{ikR} \begin{bmatrix} S_2(\Theta) & 0 \\ 0 & S_1(\Theta) \end{bmatrix} \begin{bmatrix} E_t^i \\ E_r^i \end{bmatrix} \quad [6.2]$$

Eq.[6.2] is a fundamental equation of scattered radiation by a sphere including polarization.

Mie theory solution for the scattering amplitudes:

$$S_1(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n \pi_n(\cos \Theta) + b_n \tau_n(\cos \Theta)] \quad [6.3]$$

$$S_2(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [b_n \pi_n(\cos \Theta) + a_n \tau_n(\cos \Theta)] \quad [6.4]$$

where π_n and τ_n are **Mie angular functions**

$$\pi_n(\cos \Theta) = \frac{1}{\sin(\Theta)} P_n^1(\cos \Theta) \quad [6.5]$$

$$\tau_n(\cos \Theta) = \frac{d}{d\Theta} P_n^1(\cos \Theta) \quad [6.6]$$

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:

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi r^2} P \begin{bmatrix} I_0 \\ Q_0 \\ U_0 \\ V_0 \end{bmatrix} \quad [6.7]$$

where

$$P = \begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{22} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{34} \\ 0 & 0 & P_{34} & P_{44} \end{bmatrix} \quad [6.8]$$

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.[6.7] reduces to

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi r^2} \begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{11} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{34} \\ 0 & 0 & P_{34} & P_{33} \end{bmatrix} \begin{bmatrix} I_o \\ Q_o \\ U_o \\ V_o \end{bmatrix} \quad [6.9]$$

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 (introduced in Lecture 5)

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

$$\sigma_e = \frac{4\pi}{k^2} \text{Re}[S(0^0)] \quad [6.10]$$

But for the forward direction (i.e. $\Theta = 0^0$) from Eqs.[6.3]-[6.4], we have

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

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

$$\boxed{Q_e = \frac{\sigma_e}{\pi r^2} \quad Q_s = \frac{\sigma_s}{\pi r^2} \quad Q_a = \frac{\sigma_a}{\pi r^2}} \quad [6.11]$$

where πr^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 $S_2(0^0)$).

$$Q_e = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n + 1) \operatorname{Re}[a_n + b_n] \quad [6.12]$$

$$Q_s = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n + 1) [|a_n|^2 + |b_n|^2] \quad [6.13]$$

and the absorption efficiency can be calculated as

$$Q_a = Q_e - Q_s \quad [6.14]$$

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

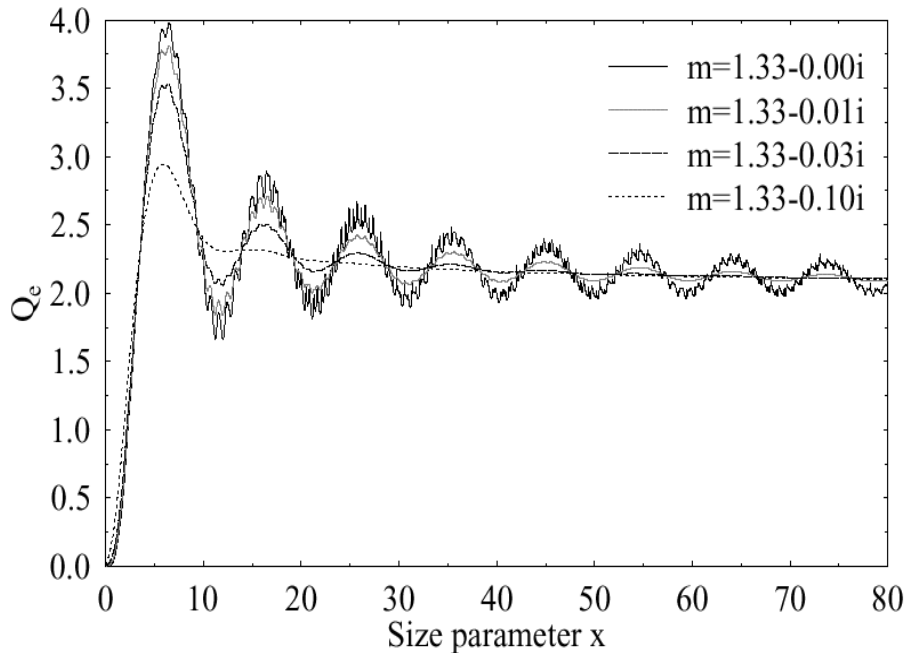


Figure 6.2 Examples of Q_e calculated with the Mie theory for several refractive indexes.

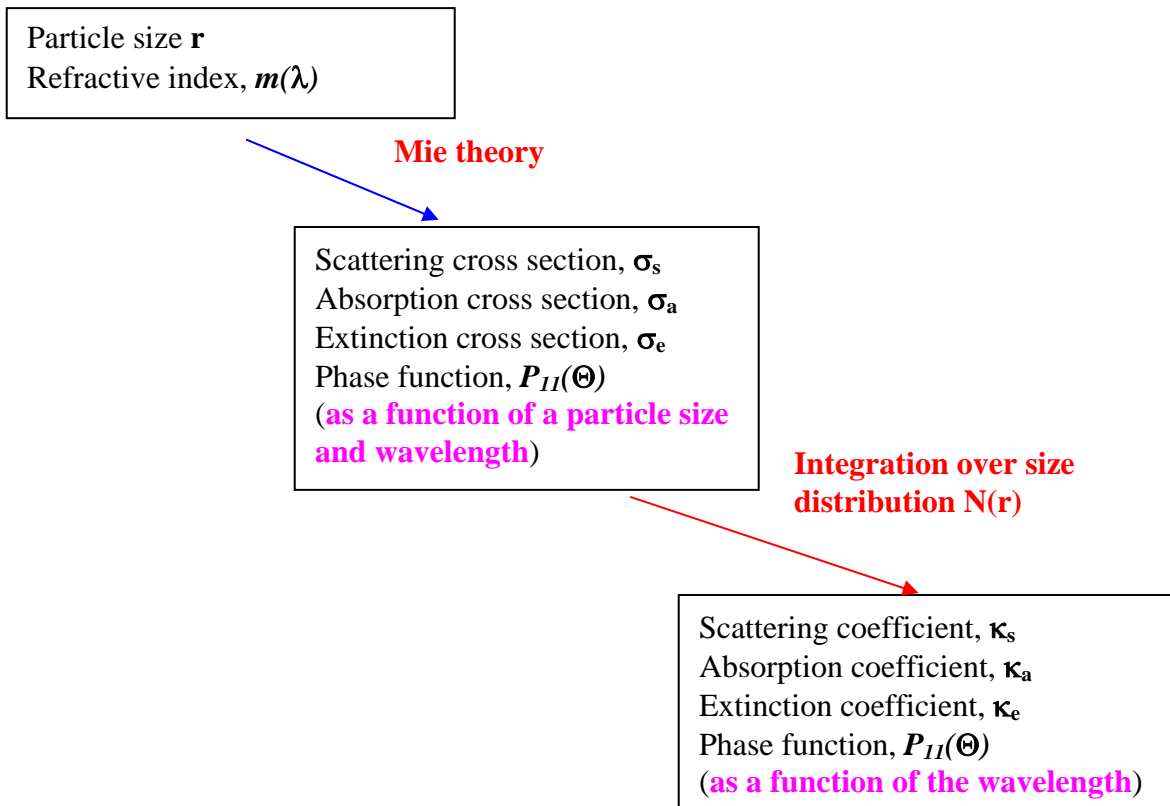
Some highlights of Mie scattering results:

- 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.6.2) 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 the Mie theory gives the extinction, scattering and absorption cross-sections (efficiency factors), the scattering amplitudes and phase matrix.

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



Integration over the particle size distribution:

For a given type of particles characterized by the size distribution $N(r)dr$, the volume extinction, scattering and absorption coefficients (in units LENGTH^{-1}) are determined as

$$k_e = \int_{r_1}^{r_2} \sigma_e(r)N(r)dr = \int_{r_1}^{r_2} \pi r^2 Q_e N(r)dr \quad [6.15]$$

$$k_s = \int_{r_1}^{r_2} \sigma_s(r)N(r)dr = \int_{r_1}^{r_2} \pi r^2 Q_s N(r)dr \quad [6.16]$$

$$k_a = \int_{r_1}^{r_2} \sigma_a(r)N(r)dr = \int_{r_1}^{r_2} \pi r^2 Q_a N(r)dr \quad [6.17]$$

Similar to Eqs.[4.11 and 4.12], the optical depth of an aerosol layer (between s_1 and s_2) is defined as

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

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

$$\omega_0 = \frac{k_s}{k_e} \quad [6.18]$$

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

No scattering: $\omega_0 = 0$

Scattering phase function of particles characterized by the size distribution $N(r)dr$

$$P(\Theta) = \frac{\int_{r_1}^{r_2} P(\Theta, r) \sigma_s N(r) dr}{\int_{r_1}^{r_2} \sigma_s N(r) dr} \quad [6.19]$$

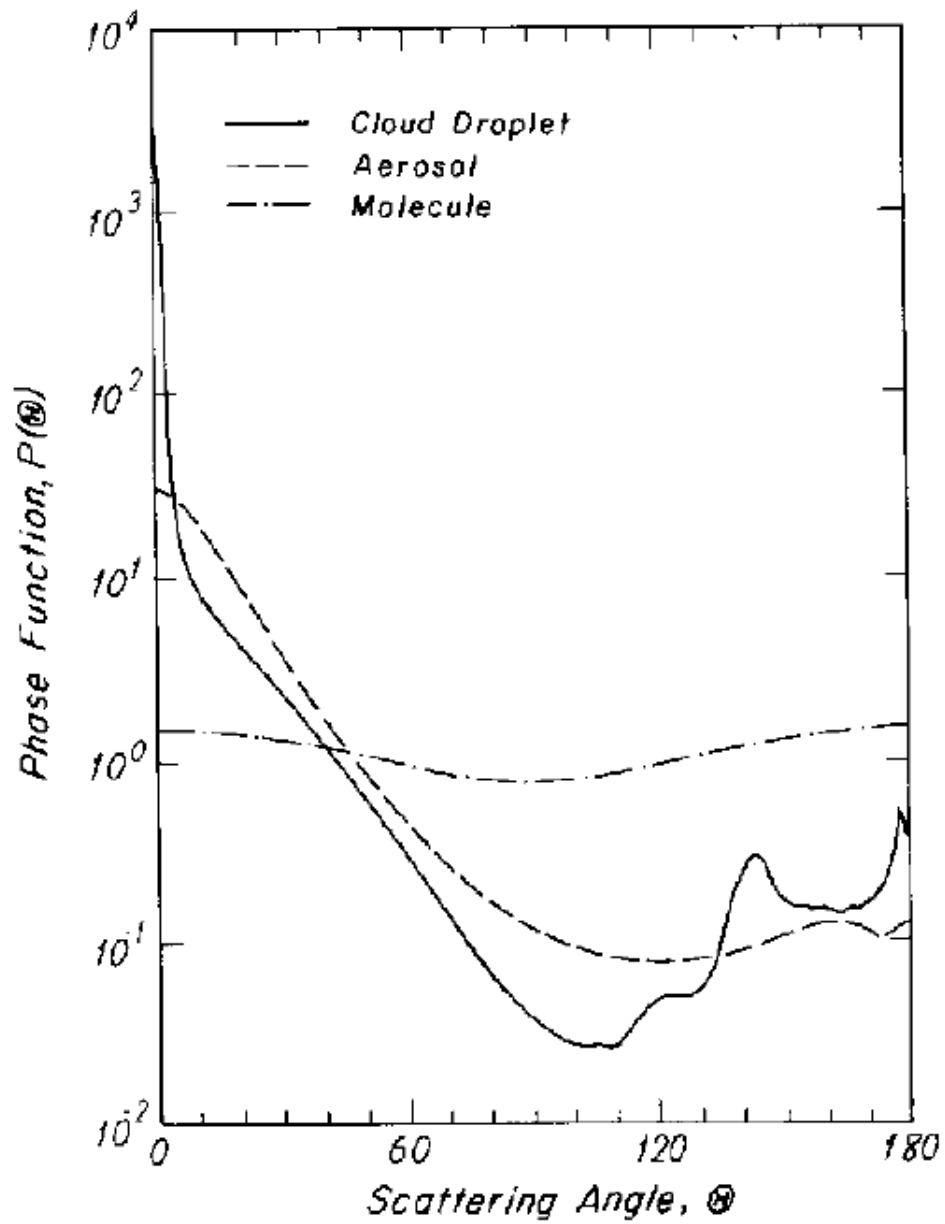


Figure 6.3 Examples of representative scattering phase functions (at a wavelength of $0.5 \mu\text{m}$) for aerosol and clouds particles. The molecular (Rayleigh) is also shown for comparison.

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

$$k_e = \sum_i k_e^i \quad k_s = \sum_i k_s^i \quad k_a = \sum_i k_a^i \quad [6.20]$$

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.[6.22-6.24] how to do it right.

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

In general, an atmospheric layer has molecules, aerosols and/or cloud particles. Thus, one needs to calculate the **effective optical properties** of this layer as an external mixture of the optical properties of these constituents.

Effective (also called total) optical depth:

$$\tau_\lambda = \tau_{a,\lambda}^M + \tau_{s,\lambda}^M + \tau_{a,\lambda}^A + \tau_{s,\lambda}^A \quad [6.21]$$

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:

$$\omega_{0,\lambda} = \frac{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A}{\tau_\lambda} \quad [6.22]$$

Effective scattering phase function:

$$P_{\lambda}(\Theta) = \frac{\tau_{s,\lambda}^M P_{\lambda}^M(\Theta) + \tau_{s,\lambda}^A P_{\lambda}^A(\Theta)}{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A} \quad [6.23]$$

Effective asymmetry parameter:

$$g_{\lambda} = \frac{\tau_{s,\lambda}^A g_{\lambda}^A}{\tau_{s,\lambda}^M + \tau_{s,\lambda}^A} \quad [6.24]$$

➤ **Optics of cloud particles:**

In contrast to atmospheric aerosols, cloud droplets consist of a single species – water.

Thus, to calculate the absorption and scattering cross sections of cloud droplets, one needs to know the size of droplets and the refractive index of water versus wavelength.

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

Extinction coefficient of cloud droplets:

Using the **effective radius**

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

and the **liquid water content (LWC):**

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

the extinction coefficient of cloud droplets is

$$k_e = \int \sigma_e(r) N(r) dr = \int Q_e \pi r^2 N(r) dr$$

and that $Q_e \approx 2$ for water droplets at solar wavelengths, we have

$$k_e \approx \frac{3}{2} \frac{LWC}{r_e \rho_w} \quad [6.25]$$

Example of cloud optical properties

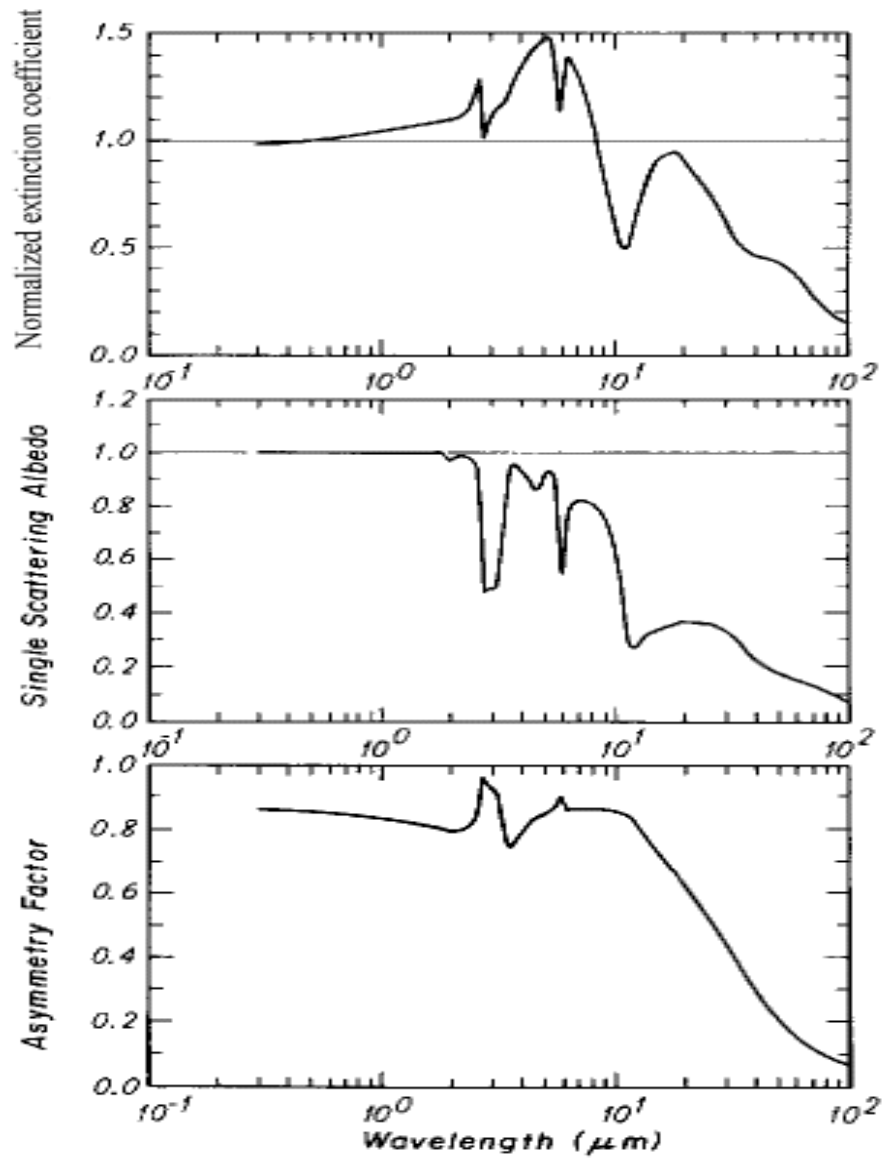


Figure 6.4 Example of optical properties as a function of wavelength of typical cumulus and stratus clouds (for a cloud droplet size distribution $r_{\text{eff}} = 6 \mu\text{m}$). Here the normalized extinction coefficient is $k_e(\lambda) / k_e(0.5 \mu\text{m})$ and $k_e(0.5 \mu\text{m}) = 42.8 \text{km}^{-1}$.

2. Main radiation law (Beer-Bouguer-Lambert 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 ΔV of infinitesimal length ds and unit area ΔA 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
$$dI_\lambda = -k_{e,\lambda} I_\lambda ds \quad [6.26a]$$

For emission:
$$dI_\lambda = k_{e,\lambda} J_\lambda ds \quad [6.26b]$$

where $\kappa_{e,\lambda}$ is the **volume extinction coefficient** (LENGTH^{-1}) and J_λ is the **source function**.

Integrating Eq.[6.26], we have

$$I_\lambda = I_{0,\lambda} \exp\left(-\int_{s_1}^{s_2} k_{e,\lambda}(s) ds\right) = I_{0,\lambda} \exp(-\tau_\lambda) \quad [6.27]$$

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

NOTE: optical depth is unitless.

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

Transmission function is defined as

$$T_\lambda = I_\lambda / I_{0,\lambda} = \exp(-\tau_\lambda) \quad [6.28]$$

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

3. Remote sensing applications based on measurements of the direct solar radiation.

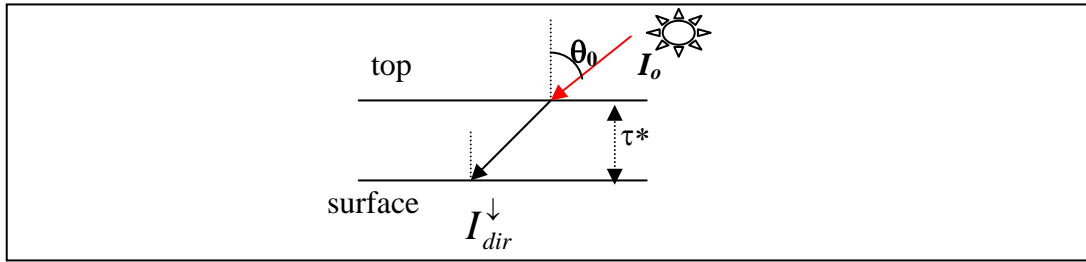
Direct radiation is a part of the radiation field that has survived the extinction passing a layer with optical depth τ and it obeys the Beer-Bouguer-Lambert law (or Extinction law):

$$I_{dir}^{\downarrow} = I_0 \exp(-\tau / \mu_0) \quad [6.29]$$

where I_0 is the incident intensity at a given wavelength at the top of a layer and μ_0 is a cosine of the incident zenith angle θ_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

$$I_{dir}^{\downarrow} = I_0 \exp(-\tau / \mu_0) \quad [6.30]$$

where τ 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 measurements:

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

$$F_{dir,\lambda}^{\downarrow} = F_{0,\lambda} \exp(-\tau_{\lambda}^* / \mu_0) \quad [6.31]$$

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

τ_{λ}^* is the optical depth of the entire atmospheric column, i.e.

$$\tau_{\lambda}^* = \tau_{\lambda}(z_{top}, 0) = \int_0^{z_{top}} k_{e,\lambda}(z) dz \quad [6.32]$$

For cloudless atmospheric conditions, τ_{λ}^* is due to attenuation by aerosols, Rayleigh scattering and gaseous absorption (O_3 and NO_2 depending on λ). Thus

$$\tau_{\lambda}^* = \tau_{a,\lambda}^M + \tau_{s,\lambda}^M + \tau_{\lambda}^A \quad [6.33]$$

From Eq.[6.31] we have

$$\ln(F_{dir,\lambda}^{\downarrow}) = \ln(F_{0,\lambda}) - \tau_{\lambda}^* / \mu_0 \quad [6.34]$$

and

$$\tau_{\lambda}^* = \mu_0 \{ \ln(F_{0,\lambda}) - \ln(F_{dir,\lambda}^{\downarrow}) \} \quad [6.35]$$

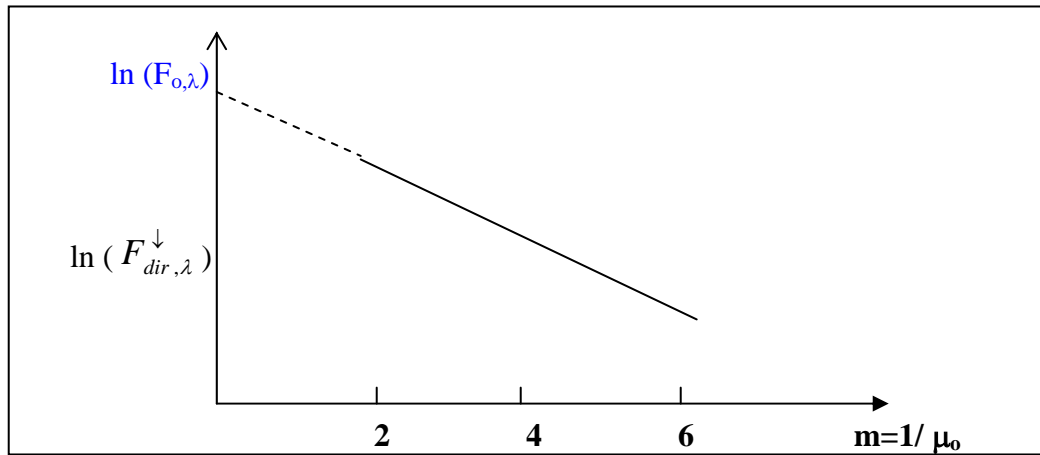
Thus

$$\tau_{\lambda}^A = \mu_0 \{ \ln(F_{0,\lambda}) - \ln(F_{dir,\lambda}^{\downarrow}) \} - \{ \tau_{a,\lambda}^M + \tau_{s,\lambda}^M \} \quad [6.36]$$

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.[6.36] enables to measure the solar flux $F_{0,\lambda}$ by plotting measured $F_{dir,\lambda}^{\downarrow}$ vs. μ_0 (called the Langley plot).



ii) If the aerosol optical depth is known or negligibly small, Eq.[6.36] gives the optical depth due to absorption of gases (used in the retrievals of O_3 and H_2O 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.

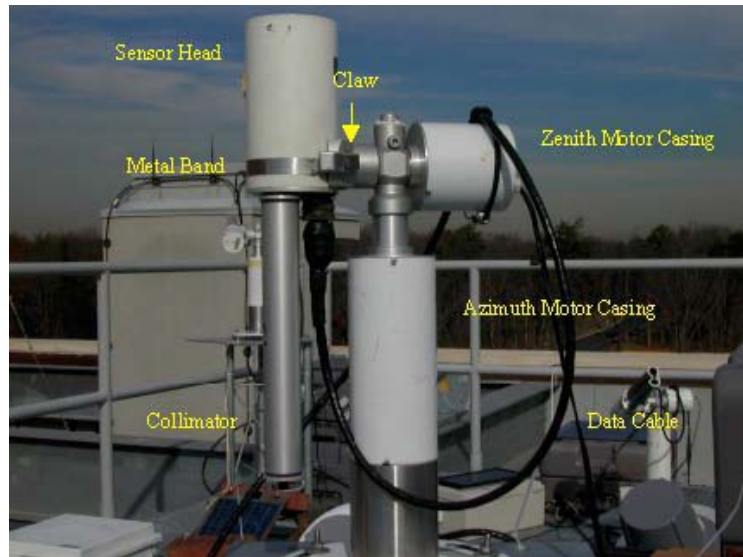


Figure 6.5 Cimel Sunphotometer operated by AERONET. Typical spectral bands (channels) of the Cimel sunptometer (central wavelengths): 340 nm, 380 nm, 440nm, 500 nm, 675 nm, 870 nm and 1020nm.

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 τ_{λ_1} and τ_{λ_2} taken at two different wavelengths λ_1 and λ_2 respectively, the Angstrom exponent α is given by

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

or

$$\frac{\tau_{\lambda_1}}{\tau_{\lambda_2}} = \left(\frac{\lambda_1}{\lambda_2} \right)^{-\alpha} \quad [6.37]$$

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