Lecture 14

Light scattering and absorption by atmospheric particulates. Part 2: Scattering and absorption by spherical particles.

Objectives:

1. Maxwell equations. Wave equation. Dielectrical constants of a medium.
2. Mie-Debye theory.
3. Optical properties of an ensemble of spherical particles.

Required Reading:

L02: 5.2

Additional/Advanced Reading:

Bohren, G.F., and D.R. Huffmn, Absorption and scattering of light by small particles. John Wiley&Sons, 1983.

Excellent web site on “The optics of a water drop: Mie scattering”
http://philiplaven.com/index1.html

1. Maxwell equations. Wave equation. Dielectrical constants of a medium.

Maxwell equations connect the five basic quantities the electric vector, $\vec{E}$, magnetic vector, $\vec{H}$, magnetic induction, $\vec{B}$, electric displacement, $\vec{D}$, and electric current density, $\vec{j}$: (in cgs system)

$$\nabla \times \vec{H} = \frac{1}{c}\frac{\partial \vec{D}}{\partial t} + \frac{4\pi}{c}\vec{j}$$

$$\nabla \times \vec{E} = \frac{-1}{c}\frac{\partial \vec{B}}{\partial t} \qquad [14.1]$$

$$\nabla \bullet \vec{D} = 4\pi\rho$$

$$\nabla \bullet \vec{B} = 0$$

where **c** is a constant (wave velocity); and **ρ** is the electric charge density.

To allow a unique determination of the electromagnetic field vectors, the Maxwell equations must be supplemented by relations which describe the behavior of substances under the influence of electromagnetic field. They are

$$\begin{aligned} \vec{j} &= \sigma \vec{E} \\ \vec{D} &= \varepsilon \vec{E} \\ \vec{B} &= \mu \vec{E} \end{aligned} \qquad [14.2]$$

where

σ is called the **specific conductivity**; **ε** is called the **dielectrical constant** (or the **permittivity**), and **μ** is called the **magnetic permeability.**

Depending on the value of σ, the substances are divided into:

conductors: $\sigma \neq 0$ (i.e., **σ is NOT negligibly small),** (for instance, metals)

***dielectrics* (or insulators):** $\sigma = 0$ (i.e., **σ is negligibly small), (for instance, air aerosol and cloud particulates)**

Let consider the propagation of EM waves in a medium which is

(a) uniform, so that **ε** has the same value at all points;

(b) isotropic, so that **ε** is independent of the direction of propagation;

(c) non-conducting (dielectric), so that **σ = 0** and therefore **j =0;**

(d) free from charge, so that **ρ** =**0.**

With these assumptions the Maxwell equations reduce to

$$\begin{aligned} \nabla \times \vec{H} &= \frac{\varepsilon}{c}\frac{\partial \vec{E}}{\partial t} \\ \nabla \times \vec{E} &= \frac{-\mu}{c}\frac{\partial \vec{H}}{\partial t} \\ \nabla \bullet \vec{E} &= 0 \\ \nabla \bullet \vec{H} &= 0 \end{aligned} \qquad [14.3]$$

Eliminating E and H in the first two equations in [14.3] and using the vector theorem, we have

$$\nabla^2 \vec{E} = \frac{\varepsilon\mu}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} \qquad [14.4]$$

$$\nabla^2 \vec{H} = \frac{\varepsilon\mu}{c^2} \frac{\partial^2 \vec{H}}{\partial t^2}$$

The above equation are standard equations of wave motion for a wave propagating with a velocity

$$v = \frac{c}{\sqrt{\varepsilon\mu}} \qquad [14.5]$$

where **c** is the speed of light in vacuum.

NOTE: for vacuum: μ = 1 and ε =1 in **cgs** units, but in SI system μ_o and ε_0 are constants such that $c = 1/\sqrt{\varepsilon_o \mu_o}$.

- For most substances (including the air) μ is unity. Thus, the electrical properties of a medium is characterized by the dielectrical constant ε.

Refractive index (or **optical constants**) of a medium is defined as

$$\boxed{m = \sqrt{\varepsilon}} \qquad [14.6]$$

assuming that μ=1.

NOTE: Strictly speaking, ε in Eq.[14.6] is the relative permittivity of medium (here it is relative to vacuum).

- ***The refractive index $m=m_r+im_i$*** in a complex number. The nonzero imaginary part m_i of the refractive index is responsible for absorption of the wave as it propagates through the medium; whereas the real part m_r of the refractive index relates to the velocity of propagation of the EM wave.
- The refractive index is a strong function of the wavelengths. Each substance has a specific spectrum of the refractive index.

- Particles of different sizes, shapes and indices of refraction will have different scattering and absorbing properties.

➢ ***<u>Refractive indices of water, ice and some aerosol species</u>***

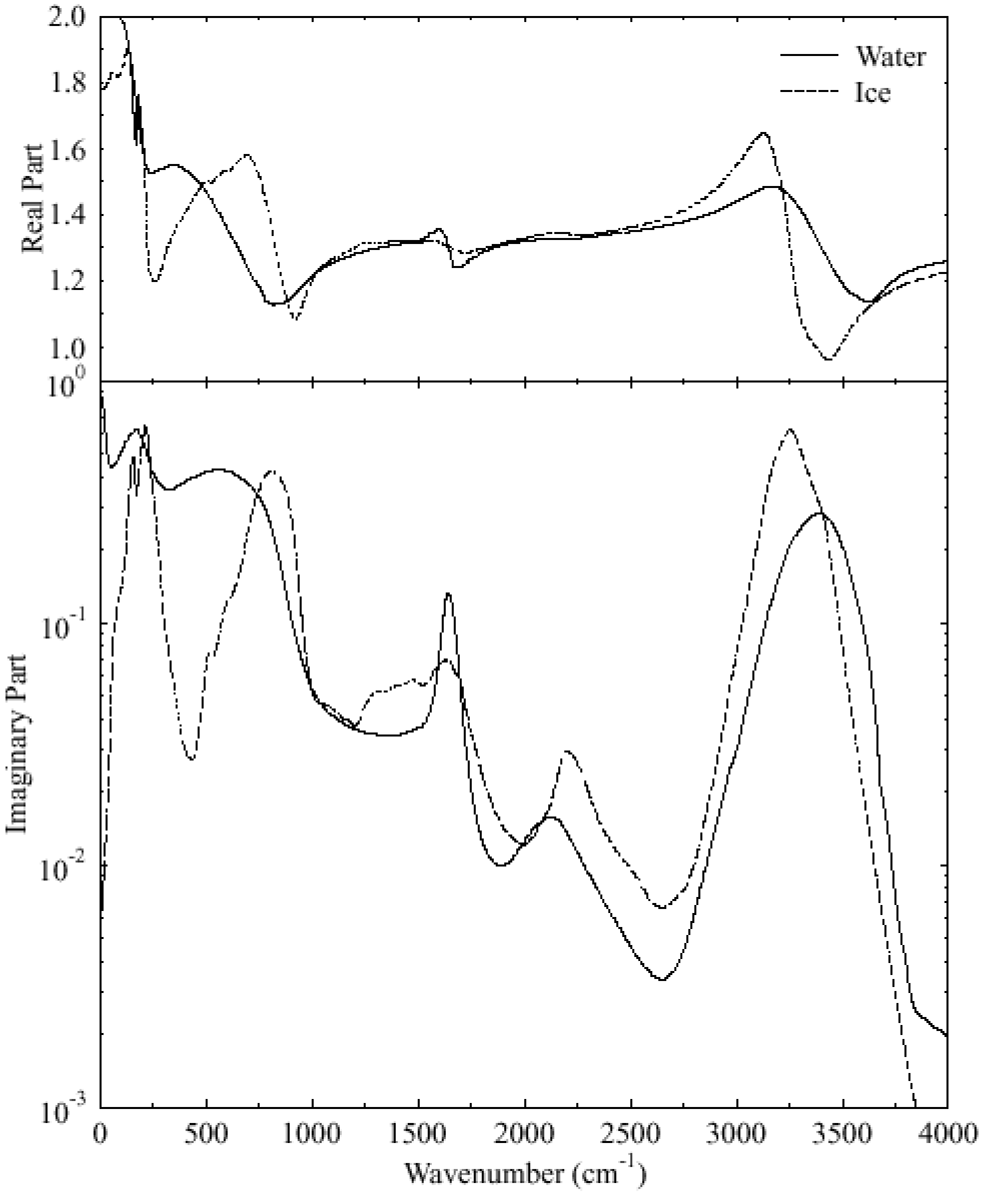

Figure 14.1 Real and imaginary part of the refractive indexes of water and ice in the IR.

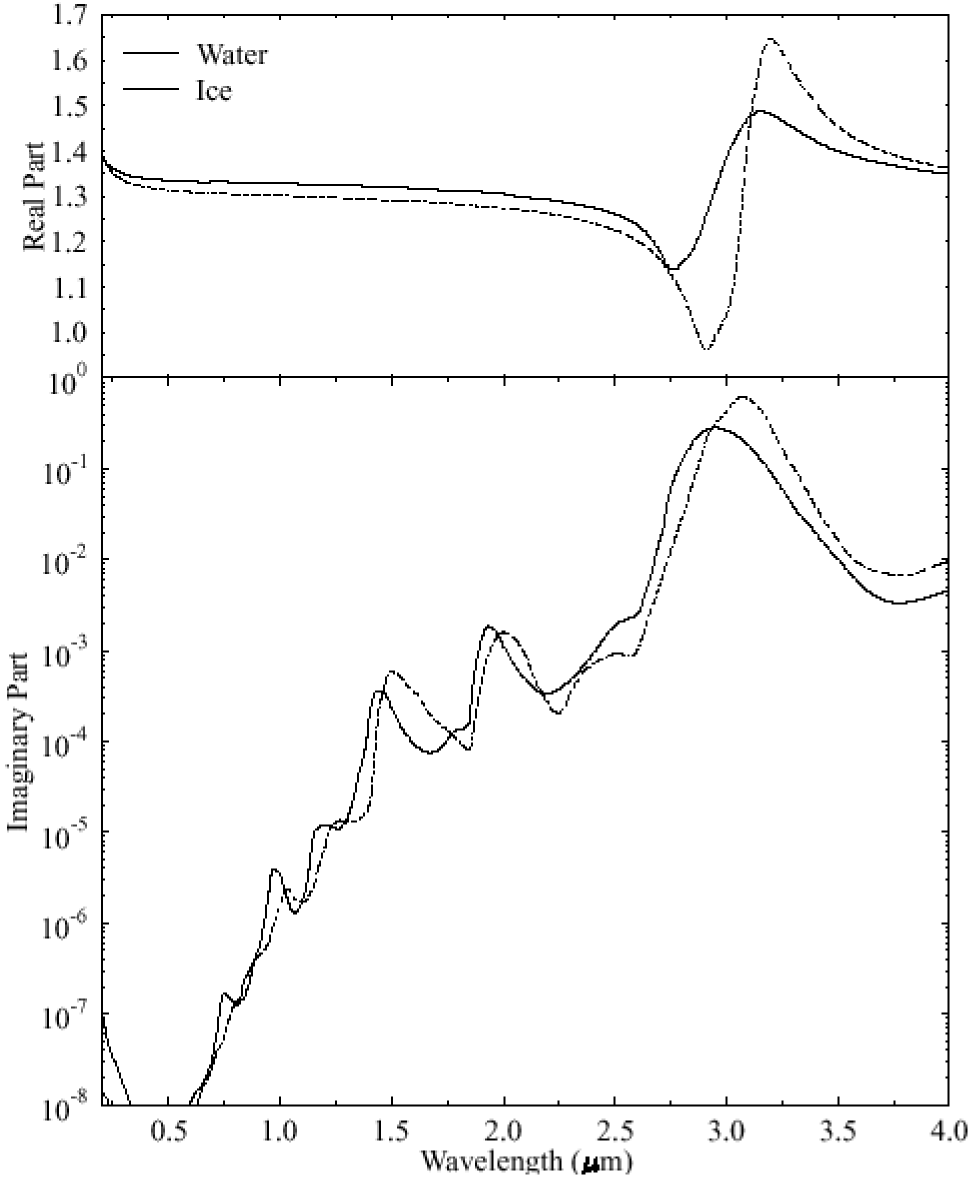

Figure 14.2 Real and imaginary part of the refractive indexes of water and ice in the visible and near-IR.

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

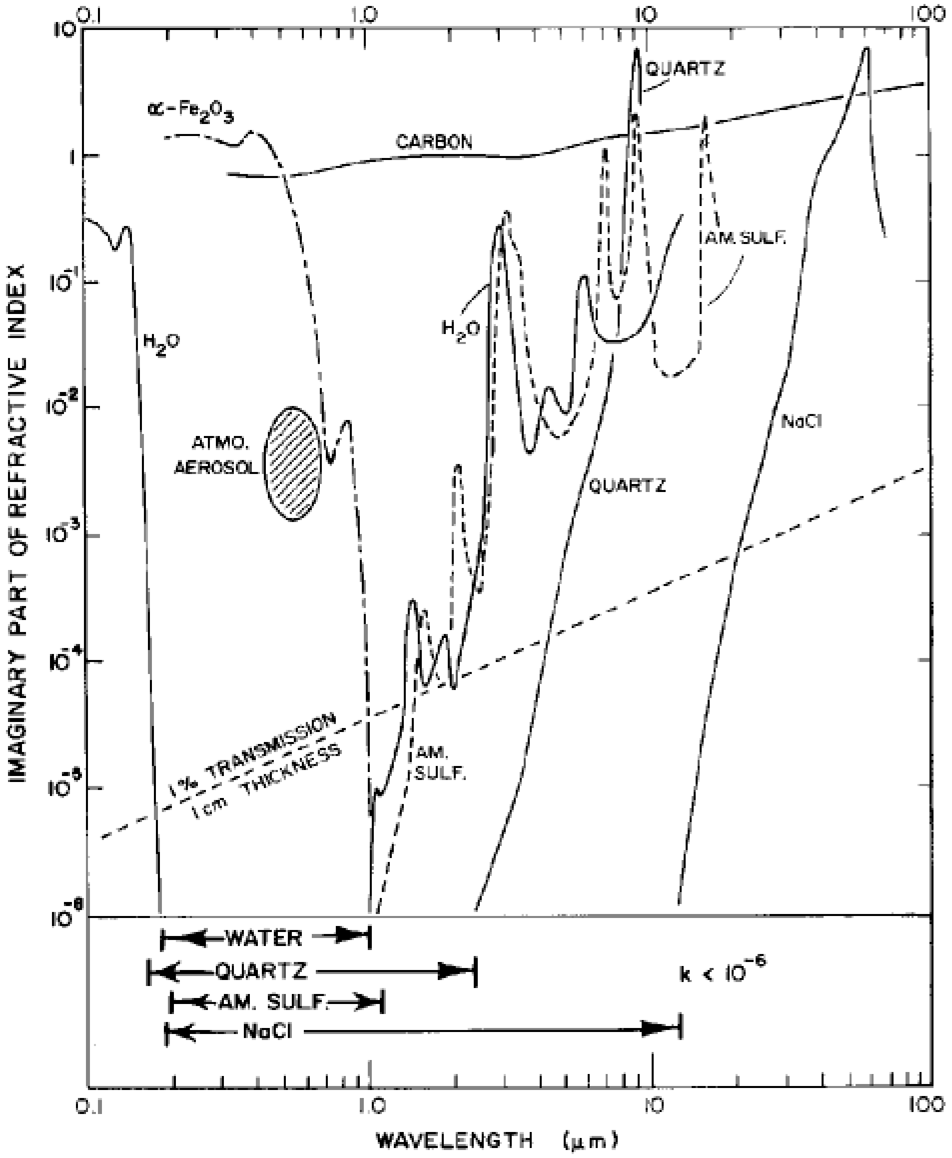

Figure 14.3 Imaginary part of the refractive indexes of some aerosol materials (Bohren and Huffman, Fig.5.16).

NOTE: Main absorbing species in the SW are black carbon (soot) and hematite (dust), but in the LW various species have high imaginary parts of the refractive index. But overall absorption (i.e., absorption coefficient) is also controlled by particle size.

- 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:

A) Volume (or mass) weighted mixing:

$$m_e = \sum_j m_j f_j \qquad [14.7]$$

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

B) Bruggeman approximation for two randomly mixed species:

$$f_1 \frac{\varepsilon_1 - \varepsilon_e}{\varepsilon_1 + 2\varepsilon_e} + f_2 \frac{\varepsilon_2 - \varepsilon_e}{\varepsilon_2 + 2\varepsilon_e} = 0 \qquad [14.8]$$

where $\boldsymbol{\varepsilon}_i$ are the dielectric constants of two materials and $\boldsymbol{f_i}$ are their volume fractions.

Recall that the refractive index is $m = \sqrt{\varepsilon}$

C) Maxwell-Garnett approximation for two specious when one is a matrix (host material) with the dielectric constant $\boldsymbol{\varepsilon}_2$ and another is an inclusion with $\boldsymbol{\varepsilon}_1$:

$$f_1 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} = \frac{\varepsilon_e - \varepsilon_2}{\varepsilon_e + 2\varepsilon_2} \qquad [14.9]$$

$$f_2 \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1} = \frac{\varepsilon_e - \varepsilon_1}{\varepsilon_e + 2\varepsilon_1}$$

NOTE: B) and C) approaches can be extended for the n-component mixtures.

➢ ***Scattering domains:***

Rayleigh scattering: 2πr/λ <<1 and *m* is arbitrary (applies to scattering by molecules and small aerosol particles);

Rayleigh-Gans scattering: $\frac{2\pi r}{\lambda}|m-1| << 1$ and $|m-1| << 1$ (not useful for atmospheric application);

Mie-Debye scattering: 2πr/λ and *m* are both arbitrary but for spheres only (applies to scattering by aerosol and cloud particles)

Geometric optics: 2πr/λ >>1 and *m* is real (applies to scattering by large cloud droplets).

2. Mie-Debye theory.

NOTE: Mie-Debye theory is often called **Mie theory** or **Lorentz-Mie theory**.

Mie theory outline:

Assumptions:

i) Particle is a **sphere** of radius ***a***;

ii) Particle is **homogeneous** (therefore it is characterized by a **single refractive index** $m = m_r + im_i$ at a given wavelength);

NOTE: Mie theory requires the relative refractive index = refractive index of a particle/refractive index of a medium. But 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, the effective refractive index must be calculated at a given wavelength.

Strategy:

1) Seek a solution of a vector wave equation (Eq.[14.4]) for $\vec{E}$ and $\vec{H}$

$$\nabla^2 \vec{E} = \frac{\varepsilon}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2}$$

with the boundary condition that the tangential component of $\vec{E}$ and $\vec{H}$ be continuous across the spherical surface of a particle. Assumption on the spherical surface of a particle allows solving the vector equation ***analytically***.

2) Re-write the wave equation in spherical coordinates and express electric field inside and outside sphere in a vector spherical harmonic expansions.

NOTE: Mie theory calculates the electromagnetic field at all points in 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 sphere):

3) Apply boundary conditions – match transverse fields at sphere surface to obtain scattered spherical wave **Mie coefficients** a_n and b_n which don't depend on the angles

but depend on size parameter **x** = **2πa/λ (*a*** is the radius of the particle) and variable **y= x *m* (*m*** is refractive index of the particle). The expression for a_n and b_n are given by Eq.[5.2.74] in L02.

4) Use series involving a_n and b_n to obtain **extinction and scattering efficiencies** (**Q_e** and **Q_s**).

5) Use series in **Mie angular functions** π_n and τ_n to obtain **scattering amplitude functions** **$S_1(\Theta)$** and **$S_2(\Theta)$,** from which the scattering phase function is derived.

Mie scattering amplitudes

(also called scattering functions) are introduced as

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

$$S_2(\Theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[b_n \pi_n(\cos\Theta) + a_n \tau_n(\cos\Theta)\right] \qquad [14.10]$$

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

$$\pi_n(\cos\Theta) = \frac{1}{\sin(\Theta)} P_n^1(\cos\Theta)$$

$$\tau_n(\cos\Theta) = \frac{d}{d\Theta} P_n^1(\cos\Theta) \qquad [14.11]$$

where P_n^1 are the associated Legendre polynomials.

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

$$\begin{bmatrix} E_l^s \\ E_r^s \end{bmatrix} = \frac{\exp(-ikr + ikz)}{ikr} \begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} \begin{bmatrix} E_l^i \\ E_r^i \end{bmatrix} \qquad [14.12]$$

Eq.[14.12] is a fundamental equation of scattered radiation including polarization in the far 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: **$S_3(\Theta) = S_4(\Theta) = 0$**

Thus, **for spheres** Eq.[14.12] reduces to

$$\begin{bmatrix} E_l^s \\ E_r^s \end{bmatrix} = \frac{\exp(-ikr+ikz)}{ikr} \begin{bmatrix} S_2 & 0 \\ 0 & S_1 \end{bmatrix} \begin{bmatrix} E_l^i \\ E_r^i \end{bmatrix} \qquad [14.13]$$

where $\exp(ikz)$ is the incident plane wave, and $\frac{\exp(-ikr)}{ikr}$ is the outgoing scattered wave.

Fundamental **extinction formula (or optical theorem)** gives the extinction cross section of a particle

$$\sigma_e = \frac{4\pi}{k^2}\mathrm{Re}[S_{1,2}(0^0)] \qquad [14.14]$$

But for the forward direction (i.e. $\Theta = 0^0$) from Eq.[14.10], we have

$$S_1(0^0) = S_2(0^0) = \frac{1}{2}\sum_{n=1}^{\infty}(2n+1)(a_n+b_n) \qquad [14.15]$$

Thus, extinction cross section is related to scattering in forward direction.

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

$$Q_e = \frac{\sigma_e}{\pi a^2} \qquad Q_s = \frac{\sigma_s}{\pi a^2} \qquad Q_a = \frac{\sigma_a}{\pi a^2} \qquad [14.16]$$

where πa^2 is the particle area projected onto the plane perpendicular to the incident beam. Mie efficiency factors are derived from the Mie scattering amplitude

$$\boxed{Q_e = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Re}[a_n+b_n]} \qquad [14.17]$$

$$\boxed{Q_s = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)[|a_n|^2+|b_n|^2]} \qquad [14.18]$$

and the absorption efficiency can be calculated as

$$\boxed{Q_a = Q_e - Q_s} \qquad [14.19]$$

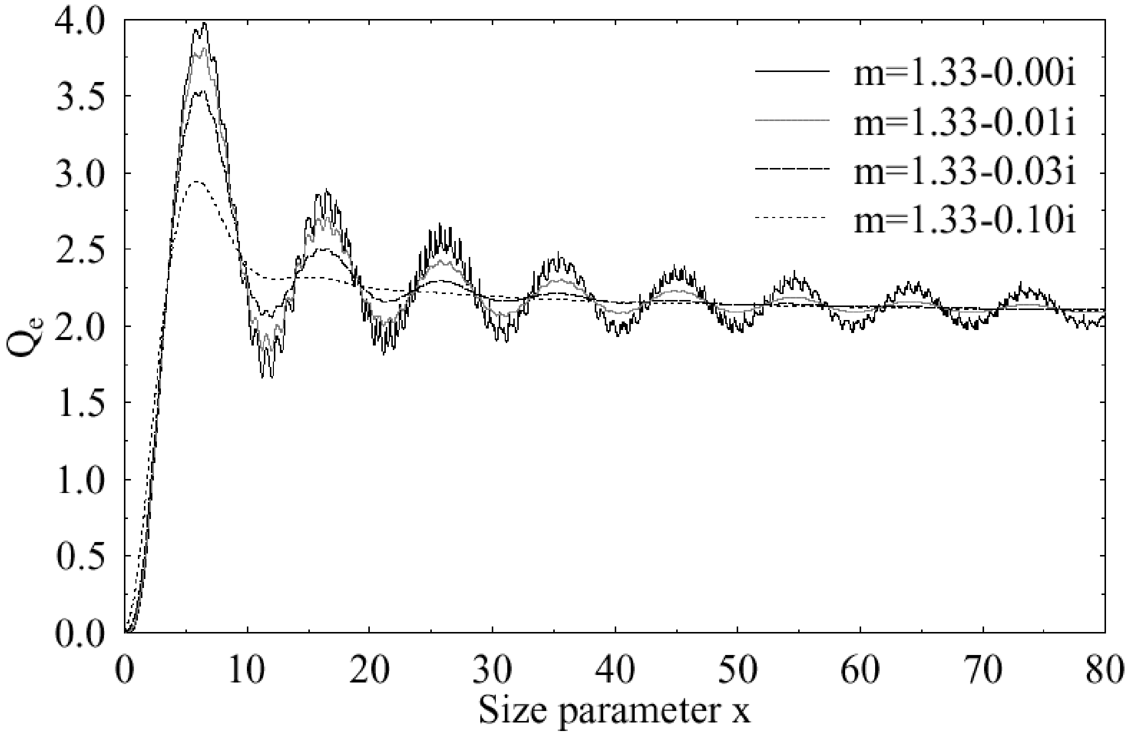

Figure 14.4 Examples of Q_e calculated with Mie theory for several refractive indexes. (more in Lab 7).

- **Scattering phase matrix**

Recall definition of Stokes parameters (see Lecture 13), which uniquely characterize the electromagnetic waves. Let $\boldsymbol{I_0}$, $\boldsymbol{Q_0}$, $\boldsymbol{U_0}$ and $\boldsymbol{V_0}$ be the Stokes parameters of incident field and I, Q, U and V be the Stokes parameters of scattered radiation

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi r^2} P \begin{bmatrix} I_o \\ Q_o \\ U_0 \\ V_o \end{bmatrix} \qquad [14.20]$$

where ***P*** is the **scattering phase matrix.**

$$P = \begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{22} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{34} \\ & & P_{34} & P_{44} \end{bmatrix} \qquad [14.21]$$

where each element depends on the scattering angle ($1/r^2$ is from solid angle)

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

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.

Thus **<u>for spheres</u>**, Eq.[14.20] reduces to

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\sigma_s}{4\pi r^2} \begin{bmatrix} P_{11} & P_{12} & & \\ P_{12} & P_{11} & & \\ & & P_{33} & -P_{34} \\ & & P_{34} & P_{33} \end{bmatrix} \begin{bmatrix} I_o \\ Q_o \\ U_0 \\ V_o \end{bmatrix} \qquad [14.22]$$

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

$$P_{11} = \frac{4\pi}{2k^2\sigma_s}\left[S_1 S_1^* + S_2 S_2^*\right]$$

$$P_{12} = \frac{4\pi}{2k^2\sigma_s}\left[S_2 S_2^* - S_1 S_1^*\right] \qquad [14.23]$$

$$P_{33} = \frac{4\pi}{2k^2\sigma_s}\left[S_2 S_1^* + S_1 S_2^*\right]$$

$$-P_{34} = \frac{4\pi}{2k^2\sigma_s}\left[S_1 S_2^* - S_2 S_1^*\right]$$

$P_{11}(\Theta) = P(\Theta)$ is the scattering phase function defined in Lecture 13.

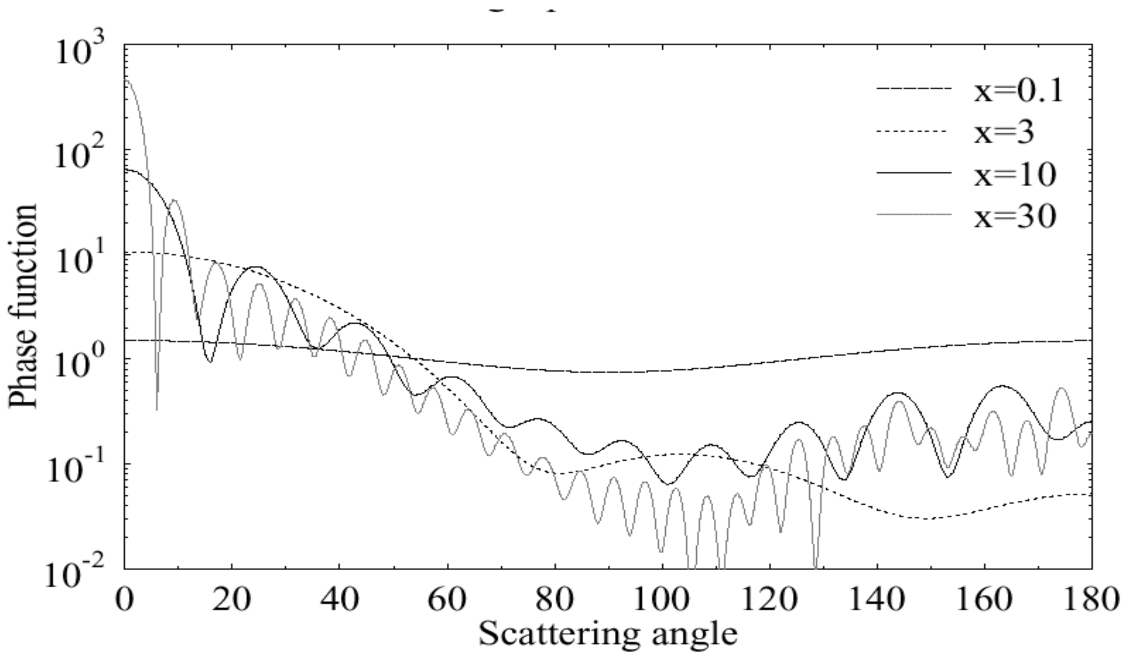

Figure 14.5 Examples of scattering phase functions calculated with Mie theory for several size parameter for nonabsorbing spheres (more in Lab 7).

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 --> 2 in geometric limit ($x \to \infty$)
  4) Oscillations (see Fig.14.4) 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 $x \to \infty$: $Q_s \to 1$ 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(Θ) increases with x.

3. Optical properties of an ensemble of spherical particles.

Mie theory gives the extinction, scattering and absorption cross-sections and the scattering phase matrix of a single spherical particle.

Recall Lecture 4 where the aerosol particle size distributions were introduced.

If the particles characterized by a size distribution N(r), the volume extinction, scattering and absorption coefficients (in units LENGTH^{-1}) are calculated as

$$\beta_e = \int_{r_{\min}}^{r_{\max}} \sigma_e(r) N(r)\, dr$$

$$\beta_s = \int_{r_{\min}}^{r_{\max}} \sigma_s(r) N(r)\, dr \qquad [14.24]$$

$$\beta_a = \int_{r_{\min}}^{r_{\max}} \sigma_a(r) N(r)\, dr$$

Single scattering albedo (unitless) if defined as

$$\boxed{\omega_0 = \frac{\beta_s}{\beta_e}} \qquad [14.25]$$

- The **single scattering albedo** gives the percentage of light which will be scattered in a single scattered event.

Scattering phase function is

$$P(\Theta) = \frac{4\pi}{2k^2\beta_s} \int_{r_{\min}}^{r_{\max}} \left[S_1 S_1^* + S_2 S_2^*\right] N(r)\, dr \qquad [14.26]$$

or

$$P(\Theta) = \frac{\int_{r_{\min}}^{r_{\max}} P_r(\Theta)\sigma_s N(r)\, dr}{\beta_s} \qquad [14.26a]$$

Asymmetry parameter is first moment of the scattering phase function and is defined as

$$\boxed{g = \frac{1}{2}\int_{-1}^{1} P(\cos\Theta)\cos(\Theta)\, d(\cos\Theta)} \qquad [14.26b]$$

g=0 for equal forward and backward; g=1 for totally forward

- 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

$$r_e = \frac{\int r^3 N(r)\,dr}{\int r^2 N(r)\,dr} \qquad [14.27]$$

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

The **liquid water content** (LWC) was defined in Lecture 4:

$$LWC = \rho_w V = \frac{4}{3}\rho_w \int \pi r^3 N(r)\,dr \qquad [14.28]$$

Using that the extinction coefficient of cloud droplets is

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

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

$$\beta_e \approx \frac{3}{2}\frac{LWC}{r_e \rho_w} \qquad [14.29]$$

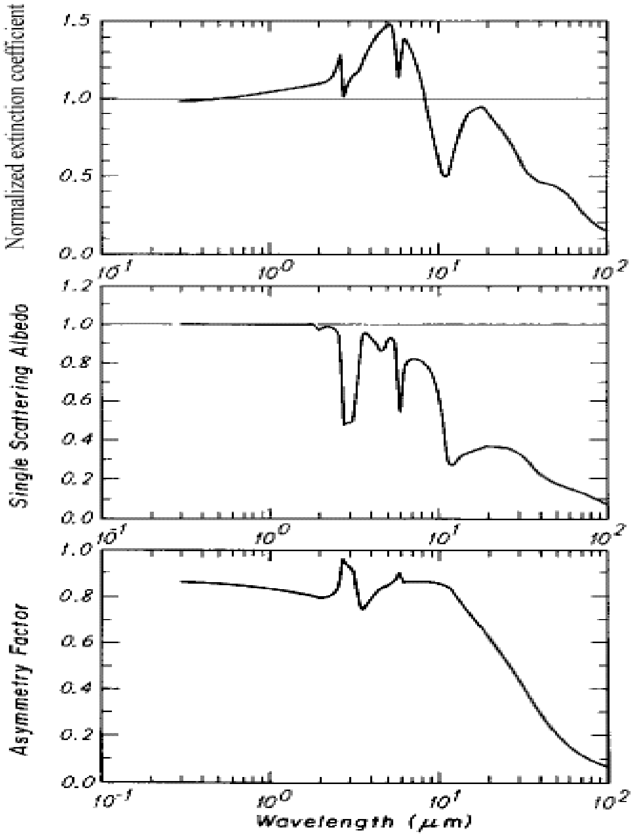

Figure 14.6 Example of optical properties of typical cumulus and stratus clouds (for a cloud droplet size distribution r_{eff} = 6 μm) . Here the normalized extinction coefficient is $\beta_e(\lambda)/\beta_e(0.5\mu m)$ and $\beta_e(0.5\mu m) = 42.8 km^{-1}$.