## Lecture 6

## Properties of atmospheric aerosols and clouds. Rayleigh scattering.

## Scattering and absorption by aerosol and cloud particles.

## Objectives:

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.
6. Scattering and absorption by aerosol and cloud particles.

## Required reading:

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

## 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. Properties of atmospheric aerosols

Atmospheric aerosols are solid or liquid particles or both suspended in air with diameters between about $0.002 \mu \mathrm{~m}$ to about $100 \mu \mathrm{~m}$.

- Interaction of the particulate matter (aerosols and clouds particles) with electromagnetic radiation is controlled by particle size, composition 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 aerosols, 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 $\left(\mathrm{SO}_{4}{ }^{2-}\right)$, nitrate $\left(\mathrm{NO}_{3}{ }^{-}\right)$, soot (elemental carbon), sea-salt ( NaCl ); minerals (e.g., quartz, $\mathrm{SiO}_{4}$ )

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)


Scanning electron microscope image of a dust particle.

## "Classical" representation of particle size spectrum:



Figure 6.1 Idealized schematic of the distribution of particle surface area of atmospheric aerosol particle (from Whitby and Cantrell, 1976).

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

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

$$
\begin{equation*}
N(r)=\sum_{i} \frac{N_{i}}{\sqrt{2 \pi} \ln \left(\sigma_{i}\right) r} \exp \left(-\frac{\ln \left(r / r_{0, i}\right)^{2}}{2 \ln \left(\sigma_{i}\right)^{2}}\right) \tag{6.1}
\end{equation*}
$$

where $\mathbf{N}(\mathbf{r})$ is the particle number concentration, $\mathbf{N}_{\mathbf{i}}$ is the total particle number concentration of i-th size mode with its median radius $\mathbf{r}_{\mathbf{0}, \mathbf{i}}$ and geometric standard deviation $\sigma_{i}$.
$k$-moment of a lognormal distribution can be found as

$$
\begin{equation*}
\int r^{k} N(r) d r=N_{0} r_{0}^{k} \exp \left(k^{2}(\ln \sigma)^{2} / 2\right) \tag{6.2}
\end{equation*}
$$

NOTE: A common approach in the satellite retrieval algorithms is to use a look-up table of aerosol candidate models.
Table 6.1 Aerosol components used in the MISR (Multi-angle ImagingSpectro
Radiometer) retrieval algorithm.

| Aerosol type | $\mathbf{r}_{0}$ | $\mathrm{r}_{\text {min }}$ | $\mathrm{r}_{\text {max }}$ | $\sigma$ | $\begin{aligned} & \text { RH } \\ & \text { (\%) } \end{aligned}$ | Vary <br> with <br> RH | $\begin{aligned} & \mathrm{H}_{\mathrm{b}} \\ & (\mathrm{~km}) \end{aligned}$ | $\begin{aligned} & \mathrm{H}_{\mathrm{t}} \\ & (\mathrm{~km}) \end{aligned}$ | $\begin{aligned} & \mathrm{H}_{\mathrm{s}} \\ & (\mathrm{~km}) \end{aligned}$ | Shape | $\begin{aligned} & \rho \\ & \mathrm{g} \mathrm{~cm}^{-3} \end{aligned}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 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 | 2.6 |
| Mineral dust (2) <br> (Accum. mode) |  |  |  |  |  |  | 5 | 10 | 10 | spheroids |  |
| Mineral dust (Coarse mode) | 1.9 | 0.5 | 15.0 | 2.6 |  |  | 0 | 2 | 10 |  |  |
| Sea salt <br> (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 | 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 $\mathrm{H}_{\mathrm{b}}$ is the base height of the aerosol layer; $\mathrm{H}_{\mathrm{t}}$ is the top height of the aerosol layer, and $H_{s}$ is the scale height of the aerosol layer; $\rho$ 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 $20 \mu \mathrm{~m}$ range.
- Cloud droplet concentration varies from about $10 \mathrm{~cm}^{-3}$ to $1000 \mathrm{~cm}^{-3}$ with average droplet concentration of a few hundred $\mathrm{cm}^{-3}$.
- The liquid water content of typical clouds, often abbreviated LWC, varies from approximately 0.05 to 3 g (water) $\mathrm{m}^{-3}$, with most of the observed values in the 0.1 to 0.3 g (water) $\mathrm{m}^{-3}$ 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 6.3 Types and properties of clouds.

| Type | Height of base (km) | Freq. over oceans (\%) | Coverage over oceans (\%) | Freq. over land (\%) | Coverage over land (\%) |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 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: |  |  |  |  |  |
| Altocumulus (Ac) | 2-7 | 46 | 22 | 35 | 21 |
| Altostratus (As) | 2-7 | (Ac+As) | ( $\mathrm{Ac}+\mathrm{As}$ ) | ( $\mathrm{Ac}+\mathrm{As}$ ) | (Ac+As) |
| High level: |  |  |  |  |  |
| Cirrus (Ci) | 7-18 | 37 | 13 | 47 | 23 |
| Cirrostratus (Cs) | 7-18 | $\mathrm{Ci}+\mathrm{Cs}+\mathrm{Cc}$ | $\mathrm{Ci}+\mathrm{Cs}+\mathrm{Cc}$ | $\mathrm{Ci}+\mathrm{Cs}+\mathrm{Cc}$ | $\mathrm{Ci}+\mathrm{Cs}+\mathrm{Cc}$ |
| 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

$$
\begin{equation*}
N(r)=\frac{N_{0}}{\Gamma(\alpha) r_{n}}\left(\frac{r}{r_{n}}\right)^{\alpha-1} \exp \left(-r / r_{n}\right) \tag{6.3}
\end{equation*}
$$

where $\mathbf{N}_{\mathbf{0}}$ is the total number of droplets $\left(\mathrm{cm}^{-3}\right) ; \mathbf{r}_{\mathbf{n}}$ in the radius that characterizes the distribution ; $\alpha$ in the variance of the distribution, and $\Gamma$ is the gamma function.

Table 6.4 Characteristics of representative size distributions of some clouds (for $\alpha=2$ )

| Cloud type | $\mathrm{N}_{\mathrm{o}}$ <br> $\left(\mathrm{cm}^{-3}\right)$ | $\mathrm{r}_{\mathrm{m}}$ <br> $(\mu \mathrm{m})$ | $\mathrm{r}_{\max }$ <br> $(\mu \mathrm{m})$ | $\mathrm{r}_{\mathrm{e}}$ <br> $(\mu \mathrm{m})$ | LWC <br> $\left(\mathrm{g} \mathrm{m}^{-3}\right)$ |
| :--- | :---: | :---: | :---: | :---: | :---: |
| Stratus: <br> over ocean <br> over land <br> Fair weather cumulus <br> $300-400$ | 10 | 6 | 15 | 17 | $0.1-0.5$ |
| Maritime cumulus | 50 | 15 | 15 | 10 | $0.1-0.5$ |
| 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

$$
\begin{equation*}
r_{e}=\frac{\int \pi r^{3} N(r) d r}{\int \pi r^{2} N(r) d r} \tag{6.4}
\end{equation*}
$$

where $\mathrm{N}(\mathrm{r})$ is the droplet size distribution (e.g., in units $\mathrm{m}^{-3} \mu \mathrm{~m}^{-1}$ ).
NOTE: Mean radius: $\mathbf{r}_{\mathbf{m}}=(\alpha+1) \mathbf{r}_{\mathbf{n}} \quad$ Effective radius: $\mathbf{r}_{\mathbf{e}}=(\alpha+3) \mathbf{r}_{\mathbf{n}}$

The liquid water content (LWC) is defined as

$$
\begin{equation*}
L W C=\rho_{w} V=\frac{4}{3} \rho_{w} \int \pi r^{3} N(r) d r \tag{6.5}
\end{equation*}
$$

## $>$ Raindrops



Nonspherical particles: shape depends of size of a rain drop

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

$$
\begin{equation*}
N(r)=N_{0} \exp (-2 \Lambda r) \tag{6.6}
\end{equation*}
$$

where $\mathrm{N}_{0}=8 \times 10^{3} \mathrm{~m}^{-3} \mathrm{~mm}^{-1}$, but, in general, $\mathrm{N}_{0}$ depends on rain type;
$\Lambda$ is related to the rainfall rate, $\operatorname{Rr}$ (in mm/hour) as $\Lambda=4.1 \mathrm{Rr}^{-0.21} \mathrm{~mm}^{-1}$

## > 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 6.2 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), $\boldsymbol{m}=\boldsymbol{n} \boldsymbol{- i k}$, is the material properties of dielectric that determines its radiative properties. In general, each material has its own spectral refractive index. The imaginary part $\boldsymbol{k}$ of the refractive index determines the absorption of the wave as it propagates through the medium; the real part $\boldsymbol{n}$ 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 6.3 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 6.4 The refractive index of water and ice in the IR.


Figure 6.5 The imaginary part of the refractive indices of some aerosol species.

- 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:

$$
\begin{equation*}
m_{e}=\sum_{j} m_{j} f_{j} \tag{6.7}
\end{equation*}
$$

where $\boldsymbol{m}_{\boldsymbol{j}}$ is the refractive index of $\boldsymbol{j}$-species and $\boldsymbol{f}_{\boldsymbol{j}}$ 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 $\mathbf{P}(\boldsymbol{\operatorname { c o s }} \Theta)$ is defined as a non-dimensional parameter to describe the angular distribution of the scattered radiation as

$$
\begin{equation*}
\frac{1}{4 \pi} \int_{\Omega} P(\cos \Theta) d \Omega=1 \tag{6.8}
\end{equation*}
$$

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

NOTE: Another form of [6.8]

$$
\begin{equation*}
\frac{1}{4 \pi} \int_{0}^{2 \pi} \int_{0}^{\pi} P(\cos \Theta) \sin \Theta d \Theta d \varphi=1 \tag{6.9}
\end{equation*}
$$

NOTE: The phase function is often expressed as

$$
P(\cos \Theta)=P\left(\theta^{\prime}, \varphi^{\prime}, \theta, \varphi^{\prime}\right)
$$

where ( $\theta^{\prime}, \varphi^{\prime}$ ) and $(\theta, \varphi)$ are the spherical coordinates of incident beam and direction of observation. Using the spherical geometry it can be shown (see S: Appendix 1) that

$$
\cos (\Theta)=\cos \left(\theta^{\prime}\right) \cos (\theta)+\sin \left(\theta^{\prime}\right) \sin (\theta) \cos \left(\varphi^{\prime}-\varphi\right)
$$

The asymmetry factor $\boldsymbol{g}$ is defined as

$$
\begin{equation*}
g=\frac{1}{2} \int_{-1}^{1} P(\cos \Theta) \cos \Theta d(\cos \Theta) \tag{6.10}
\end{equation*}
$$

$\mathbf{g}=\mathbf{0}$ for isotropic scattering

Forward scattering refers to the observations directions for which $\Theta<\pi / 2$ :
$\mathbf{g}>\mathbf{0}$ scattering in the forward direction
Backward scattering refers to the observations directions for which $\Theta>\pi / 2$ :
$\mathbf{g}<\mathbf{0}$ scattering in the backward direction

## Scattering domains:

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

Rayleigh-Gans scattering: ( $\mathbf{m} \mathbf{- 1}$ ) $\ll \mathbf{1}$ (not useful for atmospheric application)
Mie-Debye scattering: $\mathbf{2 \pi r} / \lambda$ and $m$ are both arbitrary but for spheres only (applies to scattering by aerosol and cloud particles)

Geometrical optics: $2 \pi \mathrm{r} / \lambda \gg 1$ 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, the scattering by atmospheric gases is the Rayleigh scattering.
- 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

$$
\begin{equation*}
\vec{p}_{0}=\alpha \vec{E}_{0} \tag{6.11}
\end{equation*}
$$

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 $\boldsymbol{r}$ (called far field scattering) from the dipole is given (in cgs units) by

$$
\begin{equation*}
\vec{E}=\frac{1}{c^{2}} \frac{1}{r} \frac{\partial \vec{p}}{\partial t} \sin (\gamma) \tag{6.12}
\end{equation*}
$$

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

$$
\begin{equation*}
\vec{p}=\vec{p}_{0} \exp (-i k(r-c t)) \tag{6.13}
\end{equation*}
$$

and thus the electrical field is

$$
\begin{equation*}
\vec{E}=-\vec{E}_{0} \frac{\exp (-i k(r-c t)}{r} k^{2} \alpha \sin (\gamma) \tag{6.14}
\end{equation*}
$$

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

$$
\begin{align*}
& E_{r}=-E_{0 r} \frac{\exp (-i k(r-c t)}{r} k^{2} \alpha \sin \left(\gamma_{1}\right)  \tag{6.15}\\
& E_{l}=-E_{01} \frac{\exp (-i k(r-c t)}{r} k^{2} \alpha \sin \left(\gamma_{2}\right) \tag{6.16}
\end{align*}
$$



Using that

$$
\begin{equation*}
I=\frac{1}{\Delta \Omega} \frac{c}{4 \pi}|E|^{2} \tag{6.17}
\end{equation*}
$$

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

$$
\begin{gather*}
I_{r}=I_{0 r} k^{4} \alpha^{2} / r^{2}  \tag{6.18}\\
I_{l}=I_{01} k^{4} \alpha^{2} \cos ^{2}(\Theta) / r^{2} \tag{6.19}
\end{gather*}
$$

Using that the natural light (incident beam) in not polarized ( $\boldsymbol{I}_{\mathbf{o r}}=\boldsymbol{I}_{01}=\boldsymbol{I}_{0} / \mathbf{2}$ ) and that $k=2 \pi / \lambda$, we have

$$
\begin{equation*}
I=I_{r}+I_{l}=\frac{I_{0}}{r^{2}} \alpha^{2}\left(\frac{2 \pi}{\lambda}\right)^{4} \frac{1+\cos ^{2}(\Theta)}{2} \tag{6.20}
\end{equation*}
$$

Eq.[ 6.20] gives the intensity scattered by molecules (Rayleigh scattering) for unpolarized incident light.

Rayleigh scattering phase function for incident unpolarized radiation is

$$
\begin{equation*}
P(\cos (\Theta))=\frac{3}{4}\left(1+\cos ^{2}(\Theta)\right) \tag{6.21}
\end{equation*}
$$

Eq.[ 6.20] may be rewritten in the form

$$
\begin{equation*}
I(\cos (\Theta))=\frac{I_{0}}{r^{2}} \alpha^{2} \frac{128 \pi^{5}}{3 \lambda^{4}} \frac{P(\Theta)}{4 \pi} \tag{6.22}
\end{equation*}
$$

Eq. [6.22] may be rewritten in the terms of the scattering cross section

$$
\begin{equation*}
I(\cos (\Theta))=\frac{I_{0}}{r^{2}} \sigma_{s} \frac{P(\Theta)}{4 \pi} \tag{6.23}
\end{equation*}
$$

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

$$
\begin{equation*}
\sigma_{s}=\alpha^{2} \frac{128 \pi^{5}}{3 \lambda^{4}} \tag{6.24}
\end{equation*}
$$

The polarizability is given by the Lorentz-Lorenz formula

$$
\begin{equation*}
\alpha=\frac{3}{4 \pi N_{s}}\left(\frac{m^{2}-1}{m^{2}+2}\right) \tag{6.25}
\end{equation*}
$$

where $\boldsymbol{N}_{\boldsymbol{s}}$ in the number of molecules per unit volume and $\boldsymbol{m}=\boldsymbol{n} \boldsymbol{- i k}$ in the refractive index.

For air molecules in solar spectrum: $\boldsymbol{n}$ is about 1 but depends on $\lambda$, and $\boldsymbol{k}=0$.
Thus the polarizability can be approximated as

$$
\begin{equation*}
\alpha \approx \frac{1}{4 \pi N_{s}}\left(n^{2}-1\right) \tag{6.26}
\end{equation*}
$$

Therefore, the scattering cross section of an air molecule becomes

$$
\begin{equation*}
\sigma_{s}=\frac{8 \pi^{3}\left(n^{2}-1\right)^{2}}{3 \lambda^{4} N_{s}^{2}} f(\delta) \tag{6.27}
\end{equation*}
$$

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

$$
\begin{equation*}
\tau(\lambda)=\sigma_{s}(\lambda) \int_{0}^{t o p} N(z) d z \tag{6.28}
\end{equation*}
$$

NOTE: The Rayleigh scattering cross section (Eq.[6.24]) 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:

$$
\begin{equation*}
\tau(\lambda) \approx 0.0088\left(\frac{p}{1013 m b}\right) \lambda^{-4.15+0.2 \lambda} \tag{6.29}
\end{equation*}
$$

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

$$
\begin{equation*}
L P(\Theta)=-\frac{Q}{I}=-\frac{I_{l}-I_{r}}{I_{l}+I_{r}}=-\frac{\cos ^{2} \Theta-1}{\cos ^{2} \Theta+1}=\frac{\sin ^{2} \Theta}{\cos ^{2} \Theta+1} \tag{6.30}
\end{equation*}
$$

## 6. Scattering and absorption by aerosol and cloud particles: 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;
ii) Particle is homogeneous (therefore it is characterized by a single refractive index $\boldsymbol{m}=\boldsymbol{n} \boldsymbol{- i} \boldsymbol{i}$ 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 $\boldsymbol{m}$ 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.

- Mie theory calculates the scattered 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 particle).

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

$$
\left[\begin{array}{l}
E_{l}^{s}  \tag{6.31}\\
E_{r}^{s}
\end{array}\right]=\frac{\exp (-i k r+i k z)}{i k r}\left[\begin{array}{ll}
S_{2} & S_{3} \\
S_{4} & S_{1}
\end{array}\right]\left[\begin{array}{c}
E_{l}^{i} \\
E_{r}^{i}
\end{array}\right]
$$

here $\mathrm{k}=2 \pi / \lambda, \quad E_{l}^{i}$ 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,
$\left[\begin{array}{ll}S_{2}(\Theta) & S_{3}(\Theta) \\ S_{4}(\Theta) & S_{1}(\Theta)\end{array}\right]$ is the amplitude scattering matrix (unitless)

For spheres: $\mathbf{S}_{\mathbf{3}}(\Theta)=\mathbf{S}_{\mathbf{4}}(\Theta)=\mathbf{0}$, and thus Eq.[6.31] gives

$$
\left[\begin{array}{c}
E_{l}^{s}  \tag{6.32}\\
E_{r}^{s}
\end{array}\right]=\frac{\exp (-i k r+i k z)}{i k r}\left[\begin{array}{cc}
S_{2}(\Theta) & 0 \\
0 & S_{1}(\Theta)
\end{array}\right]\left[\begin{array}{c}
E_{l}^{i} \\
E_{r}^{i}
\end{array}\right]
$$

## Eq.[6.32] is a fundamental equation of scattered radiation by a sphere including

 polarization.Mie theory scattering amplitudes:

$$
\begin{align*}
& S_{1}(\Theta)=\sum_{n=1}^{\infty} \frac{2 n+1}{n(n+1)}\left[a_{n} \pi_{n}(\cos \Theta)+b_{n} \tau_{n}(\cos \Theta)\right]  \tag{6.33}\\
& S_{2}(\Theta)=\sum_{n=1}^{\infty} \frac{2 n+1}{n(n+1)}\left[b_{n} \pi_{n}(\cos \Theta)+a_{n} \tau_{n}(\cos \Theta)\right] \tag{6.34}
\end{align*}
$$

where $\pi_{\mathbf{n}}$ and $\tau_{\mathbf{n}}$ are Mie angular functions

$$
\begin{align*}
\pi_{n}(\cos \Theta) & =\frac{1}{\sin (\Theta)} P_{n}^{1}(\cos \Theta)  \tag{6.35}\\
\tau_{n}(\cos \Theta) & =\frac{d}{d \Theta} P_{n}^{1}(\cos \Theta) \tag{6.36}
\end{align*}
$$

where $P_{n}^{1}$ are the associated Legendre polynomials.

Mie theory also gives the scattering phase matrix $\mathbf{P}(\Theta)$ that relates the Stokes parameters $\left\{\boldsymbol{I}_{\boldsymbol{0}}, \boldsymbol{Q}_{\boldsymbol{0}}, \boldsymbol{U}_{\boldsymbol{0}}\right.$ and $\left.\boldsymbol{V}_{\boldsymbol{0}}\right\}$ of incident radiation field and the Stokes parameters $\{\mathrm{I}$, Q, U and V $\}$ of scattered radiation:

$$
\left[\begin{array}{l}
I  \tag{6.37}\\
Q \\
U \\
V
\end{array}\right]=\frac{\sigma_{s}}{4 \pi r^{2}} P\left[\begin{array}{c}
I_{o} \\
Q_{o} \\
U_{0} \\
V_{o}
\end{array}\right]
$$

where

$$
P=\left[\begin{array}{cccc}
P_{11} & P_{12} & 0 & 0  \tag{6.38}\\
P_{12} & P_{22} & 0 & 0 \\
0 & 0 & P_{33} & -P_{34} \\
0 & 0 & P_{34} & P_{44}
\end{array}\right]
$$

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 for spheres, Eq.[6.37] reduces to

$$
\left[\begin{array}{l}
I  \tag{6.39}\\
Q \\
U \\
V
\end{array}\right]=\frac{\sigma_{s}}{4 \pi r^{2}}\left[\begin{array}{cccc}
P_{11} & P_{12} & 0 & 0 \\
P_{12} & P_{11} & 0 & 0 \\
0 & 0 & P_{33} & -P_{34} \\
0 & 0 & P_{34} & P_{33}
\end{array}\right]\left[\begin{array}{c}
I_{o} \\
Q_{o} \\
U_{0} \\
V_{o}
\end{array}\right]
$$

where each element of the scattering phase matrix is expressed via the scattering amplitudes $\mathbf{S}_{\mathbf{1}}(\Theta)$ and $\mathbf{S}_{\mathbf{2}}(\Theta)$

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

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

$$
\begin{equation*}
\sigma_{e}=\frac{4 \pi}{k^{2}} \operatorname{Re}\left[S\left(0^{0}\right)\right] \tag{6.40}
\end{equation*}
$$

But for the forward direction (i.e. $\Theta=0^{0}$ ) from Eqs.[6.33]-[6.34], we have

$$
S_{1}\left(0^{0}\right)=S_{2}\left(0^{0}\right)=\frac{1}{2} \sum_{n=1}^{\infty}(2 n+1)\left(a_{n}+b_{n}\right)
$$

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

$$
\begin{equation*}
Q_{e}=\frac{\sigma_{e}}{\pi r^{2}} \quad Q_{s}=\frac{\sigma_{s}}{\pi r^{2}} \quad Q_{a}=\frac{\sigma_{a}}{\pi r^{2}} \tag{6.41}
\end{equation*}
$$

where $\pi r^{2}$ is the particle area projected onto the plane perpendicular to the incident beam. Mie efficiency factors are derived from the Mie scattering amplitude

$$
\begin{align*}
& Q_{e}=\frac{2}{x^{2}} \sum_{n=1}^{\infty}(2 n+1) \operatorname{Re}\left[a_{n}+b_{n}\right]  \tag{6.42}\\
& Q_{s}=\frac{2}{x^{2}} \sum_{n=1}^{\infty}(2 n+1)\left[\left|a_{n}\right|^{2}+\left|b_{n}\right|^{2}\right] \tag{6.43}
\end{align*}
$$

and the absorption efficiency can be calculated as

$$
\begin{equation*}
Q_{a}=Q_{e}-Q_{s} \tag{6.44}
\end{equation*}
$$

- 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) $\mathrm{Q}_{\mathrm{e}}-->2$ in geometric limit $(x \rightarrow \infty)$


Figure 6.6 Examples of $\mathrm{Q}_{\mathrm{e}}$ calculated with Mie theory for several refractive indexes.

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

## Strategy to compute optics of an ensemble of spherical particles:

## Particle size $\mathbf{r}$

Refractive index, $\boldsymbol{m}(\lambda)$


Scattering cross section, $\sigma_{s}$ Absorption cross section, $\sigma_{a}$ Extinction cross section, $\sigma_{\mathbf{e}}$ Phase function, $\boldsymbol{P}_{11}(\Theta)$ (as a function of a particle size and wavelength)

Integration over size distribution $\mathrm{N}(\mathrm{r})$

Scattering coefficient, $\kappa_{s}$ Absorption coefficient, $\kappa_{a}$ Extinction coefficient, $\kappa_{\mathrm{e}}$ Phase function, $\boldsymbol{P}_{\mathbf{1 1}}(\Theta)$ (as a function of the wavelength)

## Integration over the size distribution:

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

$$
\begin{align*}
k_{e} & =\int_{r_{1}}^{r_{2}} \sigma_{e}(r) N(r) d r=\int_{r_{1}}^{r_{2}} \pi r^{2} Q_{e} N(r) d r  \tag{6.45}\\
k_{s} & =\int_{r_{1}}^{r_{2}} \sigma_{s}(r) N(r) d r=\int_{r_{1}}^{r_{2}} \pi r^{2} Q_{s} N(r) d r  \tag{6.46}\\
k_{a} & =\int_{r_{1}}^{r_{2}} \sigma_{a}(r) N(r) d r=\int_{r_{1}}^{r_{2}} \pi r^{2} Q_{a} N(r) d r \tag{6.47}
\end{align*}
$$

## Mass coefficients $=$ volume coefficients/particle mass concentration, $\mathbf{M}$

Similar to Eq.[5.13], the optical depth of an aerosol layer is given as

$$
\tau_{\lambda}\left(s_{1}, s_{2}\right)=\int_{s_{1}}^{s_{2}} k_{e, \lambda} d s=\int_{s_{1}}^{s_{2}} M k_{m, e, \lambda} d s
$$

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

$$
\begin{equation*}
\omega_{0}=\frac{k_{s}}{k_{e}} \tag{6.48}
\end{equation*}
$$

NOTE: No absorption (conservative scattering): $\omega_{0}=1$
No scattering: $\omega_{0}=0$

Scattering phase function of particles characterized by the size distribution $\mathbf{N}(\mathbf{r})$

$$
\begin{equation*}
P(\Theta)=\frac{\int_{r_{1}}^{r_{2}} P(\Theta, r) \sigma_{s} N(r) d r}{\int_{r_{1}}^{r_{2}} \sigma_{s} N(r) d r} \tag{6.49}
\end{equation*}
$$



Figure 6.7 Examples of scattering phase functions at a wavelength of $0.5 \mu \mathrm{~m}$.
$>$ Optical properties of the external mixture (i.e., the mixture of several types of particles)

$$
\begin{equation*}
k_{e}=\sum_{i} k_{e}^{i} \quad k_{s}=\sum_{i} k_{s}^{i} \quad k_{a}=\sum_{i} k_{a}^{i} \tag{6.50}
\end{equation*}
$$

where $k_{e}^{i}, k_{s}^{i}$ and $k_{a}^{i}$ are calculated for each particle type characterized by its particle size distribution $\mathrm{N}_{\mathrm{i}}\left(\mathrm{r}\right.$ ) and a refractive index (or effective refractive index) $\mathrm{m}_{\mathrm{i}}$.

NOTE: Do not sum the single scattering albedo and scattering phase functions !!!

- 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.


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

Effective (also called total) optical depth:

$$
\begin{equation*}
\tau_{\lambda}=\tau_{a, \lambda}^{M}+\tau_{s, \lambda}^{M}+\tau_{a, \lambda}^{A}+\tau_{s, \lambda}^{A} \tag{6.51}
\end{equation*}
$$

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:

$$
\begin{equation*}
\omega_{0, \lambda}=\frac{\tau_{s, \lambda}^{M}+\tau_{s, \lambda}^{A}}{\tau_{\lambda}} \tag{6.52}
\end{equation*}
$$

## Effective scattering phase function:

$$
\begin{equation*}
P_{\lambda}(\Theta)=\frac{\tau_{s, \lambda}^{M} P_{\lambda}^{M}(\Theta)+\tau_{s, \lambda}^{A} P_{\lambda}^{A}(\Theta)}{\tau_{s, \lambda}^{M}+\tau_{s, \lambda}^{A}} \tag{6.53}
\end{equation*}
$$

## Effective asymmetry parameter:

$$
\begin{equation*}
g_{\lambda}=\frac{\tau_{s, \lambda}^{A} g_{\lambda}^{A}}{\tau_{s, \lambda}^{M}+\tau_{s, \lambda}^{A}} \tag{6.54}
\end{equation*}
$$

## Optics of cloud particles

- In contrast to atmospheric aerosols, the 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 $20 \mu \mathrm{~m}$ range.


## Extinction coefficient of cloud droplets:

Using the effective radius

$$
r_{e}=\frac{\int \pi r^{3} N(r) d r}{\int \pi r^{2} N(r) d r}
$$

and the liquid water content (LWC):

$$
L W C=\rho_{w} V=\frac{4}{3} \rho_{w} \int \pi r^{3} N(r) d r
$$

the extinction coefficient of cloud droplets is

$$
k_{e}=\int \sigma_{e}(r) N(r) d r=\int Q_{e} \pi r^{2} N(r) d r
$$

and that $Q_{e} \approx 2$ for water droplets at solar wavelengths, we have

$$
\begin{equation*}
k_{e} \approx \frac{3}{2} \frac{L W C}{r_{e} \rho_{w}} \tag{6.55}
\end{equation*}
$$

## Example of cloud optical properties



Figure 6.8 Example of optical properties of typical cumulus and stratus clouds (for a cloud droplet size distribution $\mathrm{r}_{\text {eff }}=6 \mu \mathrm{~m}$ ). Here the normalized extinction coefficient is $k_{e}(\lambda) / k_{e}(0.5 \mu \mathrm{~m})$ and $k_{e}(0.5 \mu \mathrm{~m})=42.8 \mathrm{~km}^{-1}$.

