

MODELING INTERACTION OF RADIATION WITH AEROSOLS USING MIE SCATTERING FORMULATION AND RETRIEVAL OF AEROSOL SIZE PARAMETERS

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By

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To my family

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Abstract

Determining the aerosol size distribution function is a fundamental problem in aerosol science. The size distribution function describes how certain property of a particle population is distributed over different particle sizes. Since the size distribution cannot be measured directly, an estimate for the size distribution is determined from indirect measurements using inverse theory. Estimating size distribution is an ill-posed inverse problem, therefore, we have developed non-linear inversion algorithm to retrieve aerosol size parameters. The non-linear inversion algorithm has been applied to measured aerosol optical depth at Bahrain station, which is one of the stations in Aerosol Robotic Network (AERONET).

The sensitivity of the retrieved columnar size distribution to the assumed refractive index was investigated for aerosol containing water droplets and dust. The dust aerosol shows insignificant sensitivity to changes in the imaginary part of the refractive index while water soluble aerosols have shown appreciable sensitivity to the imaginary part of the refractive index.

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Introduction

Atmospheric aerosols affects the Earth's energy budget mainly through two factors. One is a direct effect in which the aerosol particles scatter and absorb the solar and thermal radiation. The second is an indirect effect in which they change the microphysical and optical properties of cloud droplets acting as cloud condensation nuclei [1]. But the quantitative assessment of the radiative energy budget of the atmosphere is uncertain because of lack of knowledge of radiative impacts of aerosol as well as the optical properties of aerosols [2]. Monitoring of atmospheric aerosol is fundamentally a difficult problem. First compared to atmospheric gases, aerosol is highly inhomogeneous and variable, that is, aerosol observation have to be global and continuous. Second, the available accuracy of aerosol characterization is often not sufficient [3,4].

The difficulties in accessing the contribution of aerosols to radiative processes are caused by incomplete knowledge of aerosol macrophysical properties (sources, sinks, and loading) and of aerosol microphysical properties (composition, size distribution, chemical interaction, lifetime, and diurnal variation). The discrete spatial and temporal nature of both natural (e.g., volcanic eruption, wind-lofted (e.g., Saharan dust, and sea spray) and anthropogenic aerosol injection (e.g., biomass burning and industrial pollution) makes the problem particularly challenging [3].

Aerosol size distributions describe the number of particles observed in a certain radius size ranges. For convenience the entire aerosols size ranges are divided into three major categories. These are the nucleation mode containing particles of size less than $0.1 \mu m$, the accumulation mode particles having size between $0.1 \mu m$ and $1 \mu m$ and the coarse mode particles of size greater than $1.0 \mu m$. The nucleation mode particles are the secondary particles formed from gas to particle conversion mechanism and the accumulation mode particles are formed mostly by the coagulation of two or more nucleation particles. The coarse mode particles are the primary particles that are directly injected into the atmosphere such as, sea-salt particles over the ocean and fine sand particles and biogenic dust over the land.

Modelling the aerosol effects on atmospheric radiation, requires the radiative transfer equation which solves the aerosols optical properties, thus aerosol optical properties. Aerosol optical thickness $\tau(\lambda)$ (loading), phase function $P(\theta, \lambda)$ (angular dependence of light scattering) and the single scattering albedo $\omega(\lambda)$ (ratio of scattering to extinction). The sensitivity of radiative forcing to observe natural and anthropogenic variations of the above listed optical properties characterizes the impact of the atmospheric aerosol on climate change [5].

A large fraction of the aerosol mass present in the earth's atmosphere is irregularly shaped mineral dust. It has been shown in various studies that neglecting the nonsphericity of mineral aerosol particles can yield large errors in simulated radiances and retrieved aerosol parameters. Light scattering and absorption by nonspherical particles or particles with surface irregularities depend not only the incident wavelength and the refractive index of the particle, but also on the particle size, shape,

orientation and surface deformation. To model accurately the light scattering and absorption by natural particles, one usually needs to build mathematical models to describe the particle shapes and use appropriate light scattering models to calculate the single scattering properties for these specific particle shapes [6].

The aerosol optical depth is a very valuable aerosol characteristic, which is a key parameter for various aerosol related studies, such as radiative forcing atmospheric corrections of the aerosol effect on the remote sensing etc. By monitoring size distribution function in different situations and circumstances it is possible to obtain information on formation, behavior and effects of aerosol particles. Determining the aerosol size distribution from forward spectral extinction measurements requires the inversion of a Fredholm integral equation of the first kind [7-9]. This equation gives the extinction as an integral over the size distribution with a kernel determined by scattering theory.

In this thesis, we use the Mie code which is developed by M.Mishenko which is available on the world wide web at <http://www.giss.nasa.gov/crmim.book> [6]. We have developed a retrieval algorithms to be used along with Mie code to derive aerosol size distribution from measured extinction data.

The rest of the thesis is organized as follows: In chapter one, we present atmospheric aerosol definitions, radiative and climatic effects of aerosols, interaction of electromagnetic radiation with aerosols, and the key parameters that govern scattering and absorption of electromagnetic radiation with aerosols.

In chapter two, we present Mie scattering, electromagnetic wave equation and its solution, and the far field solutions and extinction parameters.

In chapter three, we present, the inverse problem, retrieval formalism, retrieval of aerosol size distributions from optical thickness measurements.

In chapter four, we present, simulation of extinction efficiency by spherical particles, Ground-based network, sensors functionality at AERONET, extinction data, real and simulated extinction data.

In chapter five, we present the results and discussion. Finally, in chapter six we present the conclusion.

Chapter 1

Atmospheric aerosols

In this chapter, we review briefly some important aspects of tropospheric aerosols, including their sources, radiative and climatic impacts of aerosols, interaction of electromagnetic radiation with aerosols and the key parameters that govern scattering and absorption of aerosols.

1.1 What are atmospheric aerosols?

Atmospheric aerosols are mixtures of solid or liquid particles which are suspended in the atmosphere. Particles are referred to as aerosols as long as they are suspended in the atmosphere. They are in micron and sub-micron ranges. This definition encompasses a broad array of different substances with widely varying shapes, sizes, sources, sinks, and physical and chemical properties. As their life times are on the order of days, aerosols are inhomogeneous in time and in space, with much higher concentrations near sources. Thus properties are extremely difficult to characterize aerosols on the global scale. Atmospheric aerosols can originate from natural and man made (anthropogenic) source [4]. Particles such as mineral aerosols, sea salt, volcanic eruption which are directly injected from the atmosphere are known as primary particles.

Where as particles released from precursor gases are known as secondary aerosols e.g sulfates.

1.2 Radiative and Climatic Effects of Aerosols

Aerosol particles play an important role in radiative budget of the atmosphere. The earth's radiation budget is the balance between incoming energy from the sun and outgoing energy from the earth. If the Earth and Earth's atmosphere retain more solar energy than it radiates back to space, the earth will warm. If the earth and the earth system radiates more energy to space than it receives from the sun, the earth will cool. The radiative properties of particles in the atmosphere can be expressed by its complex refractive index and it is designated by $m = m_r + im_i$, where the real part represents the scattering whereas the imaginary part represents absorption. The refractive index is strongly dependent on the chemical composition of the particles. The chemical composition and hence refractive index depends on the source of particles. The real part of the refractive index in the visible spectrum is not very sensitive to the chemical composition. The real refractive index is variable in the range between $n=1.3-1.65$ for most aerosol types and relative humidity and the imaginary part of the index is much more variable, ranging from $1E-9$ (e.g water droplets) to more than 0.1 in very sooty or iron-rich dust aerosols [7].

Atmospheric aerosols exert both direct and indirect climate forcing. They directly interact with solar and thermal radiation and indirectly they affect either the amount or the radiative properties of other components of the system, such as cloud and hence produce an additional radiative forcing. The magnitude of radiative direct radiative forcing by aerosols at any location and time depends on the amount of

aerosols present, their optical properties, underlying surface albedo and the position of the sun. The aerosols have potential to warm or cool the earth-atmosphere system depending upon their absorptivity and surface albedo. The scattering and absorption properties of individual particles depend upon the particle shape, size, refractive index and wavelength of incident radiation. The radiative properties of an aerosol layer depend on the above factors as well as on the spatial distribution of the particles, angle of incidence of radiation, and for the nonspheric particles, on their orientation. However, the impact aerosol layer is not only determined by particle size distribution and composition but also on its locations and thickness of the atmosphere, the nature of the underlying surface and the presence of clouds. The main difference between the radiative forcing by greenhouse gases and aerosols is that direct greenhouse gases forcing is both during day and night time and applicable for cloudy and cloud free conditions. But the aerosols direct forcing is restricted to only daytime conditions and is highest in cloud free conditions. Also, most of the greenhouse gases have very long lifetime compared to aerosols and hence they are well mixed and forcing is more or less uniform throughout the world. Aerosol lifetime is very short compared to greenhouse gases and hence the aerosol forcing is limited near to the aerosol sources. Typical atmospheric aerosols, which play a crucial role, are of the order of $1\ \mu m$ in radius or less and are mostly nonabsorbent at visible wavelengths.

These aerosols produce a cooling of the global surface temperature because of reduced solar heating due to radiation being backscattered to the space. But the aerosols like soot and desert dust absorb sufficiently large amount of sunlight, which can lead to warming of the global climate. Some of the aerosols also absorb and reemit at thermal wavelengths that contribute a small, but not negligible amount. Also atmospheric

aerosols are found to affect indirectly the amount, type and distribution of clouds. An increase in aerosol number concentration will result in an increase in the number of cloud condensation nuclei, which leads to clouds with higher number of smaller size cloud droplets. The assessment of radiative effects of aerosols, especially of the indirect effect on the radiative balance of the Earth are highly uncertain [5,9]. The uncertainty arises due to the difference in concentration and evaluation of the cloud droplets number concentration. On the other hand, the uncertainty in the estimates of indirect radiative forcing results from the complexity of the relationship between the aerosol mass input to the atmosphere and both the radiative properties of clouds and cloud dynamics [9]. An increase in the number of cloud droplets enhances multiple scattering of light within the clouds, which increases the optical depth and albedo of the cloud. This effect of aerosols on clouds is in general termed as the indirect effect. Stratospheric aerosols scatter visible solar radiation back to the space, leading to a cooling of the Earth's surface. They also absorb upwelling long wave radiation, warming the stratospheric layers where particles reside and thus provide a significant forcing of the climate.

1.3 Interaction of Electromagnetic Radiation With Aerosols

Scattering of sunlight by molecules and particles takes place in the atmosphere. Scattering is the process by which molecules and particles in the path of an electromagnetic waves continuously extracts the energy from the incident wave and reradiates that same energy in all directions. The two most scattering process in the atmosphere are the elastic scattering, in which the wavelength of the scattered radiation is the

same as that of the wavelength of incident radiation. Inelastic scattering, in which the wavelength of the scattered radiation is different from that of the incident radiation. The elastic scattering can also be classified into Rayleigh scattering, and Mie scattering depending on the size of the particle as compared to the wavelength of the incident radiation. Inelastic scattering, in which the wavelength of the scattered radiation is different from that of the incident radiation. When the particle size is much smaller than the incident wavelength ($< 0.03 \lambda$), the scattering is called Rayleigh scattering. Since the photon energies of the scattered photons is not changed Rayleigh scattering considered as elastic scattering is inversely proportional to the fourth power of the wavelength and symmetric in the forward and backward directions (Rayleigh scattering).

Scattering can be explained in terms of the electromagnetic theory. The electric field of the incident wave sets in oscillation of the electric charges in the particle. When the size of the particle is small than the wavelength of the incident radiation an electric dipole is set up and the scattering is symmetric because an electric dipole radiates equal amount of fluxes in both forward and backward directions (Rayleigh scattering).

Mie scattering of particles in which the size of particles are comparable to the incident wavelength of the incident radiation. This scattering produces a pattern like an antenna lobe, with a sharper and more intense forward lobe for larger particles. Rayleigh scattering is a special case of Mie scattering. In Mie scattering a forward peak is developed and as the particle size increases the scattering is no more symmetric and more and more radiation is scattered in the forward direction than in the backward direction. The intensity of scattered radiation in case of Rayleigh scattering

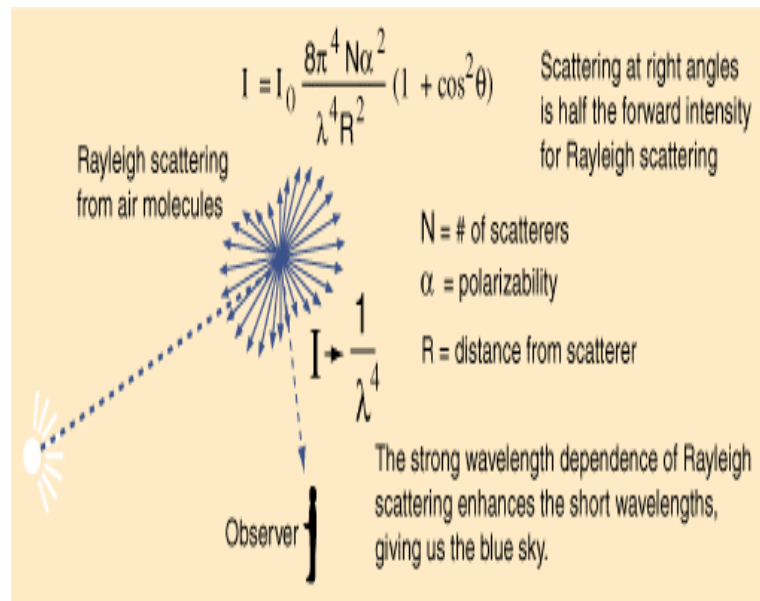


Figure 1.1: Rayleigh scattering by air molecules in the atmosphere

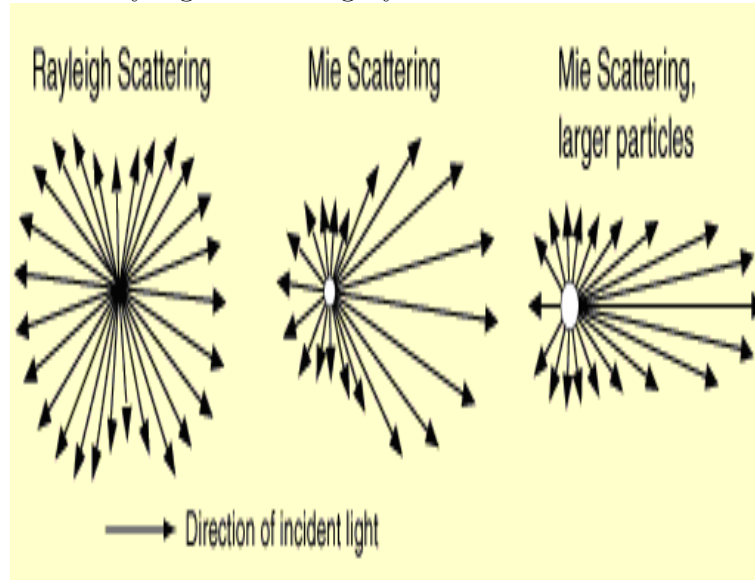


Figure 1.2: Mie scattering by aerosol particles in the atmosphere

is given by [11]

$$I_S = |E|^2 = \frac{\omega^4}{c^4 r^2} \alpha^2 (1 + \cos^2(\theta)) I_0^2$$

Where θ =scattering angle, ω frequency of incident radiation, c velocity of light, r distance between scatterer and the point under observation, α polarizability of the scatterer.

The important characteristics of Rayleigh scattering are:

1. The scattered intensity is proportional to the fourth power of frequency of incidence radiation.
2. The scattered intensity is inversely proportional to the square of the distance between the molecule and the point of observation.
3. The angular distribution of the scattered intensity is symmetric in the forward and backward directions and is given by $(1 + \cos^2\theta)$:

The important characteristics of Mie scattering are:

1. The angular distribution of the scattered radiation intensity is complicated and the complexity increases with increasing particle size.
2. The ratio of energy scattered in the forward direction increases with increasing particle size.
3. The dependence of scattering on wavelength decreases with increasing particle size.

1.4 The Key parameters that govern the scattering and absorption of Electromagnetic Radiation

(A) Size Parameter: The ratio of the circumference of the particle to the wavelength is known as size parameter and it is denoted by x . When the size parameter, x exceeds unity the scattering diagram begins to develop peak and troughs.

$$x = \frac{2\pi r}{\lambda}$$

(B) Extinction coefficient: The fraction of energy removed per unit path length from an incident wave with energy flux density 1 by a collection of aerosol particles characterized by the particle size distribution is known as extinction coefficient and is denoted by β_{ext} .

$$\beta_{ext} = \beta_{sca} + \beta_{abs}$$

(C) Single scattering albedo: The probability of a photon that interacts with particle and reappears as scattered radiation and it is denoted by ω_0 : For a nonabsorbing aerosols $\omega_0=1$ and for a perfectly absorbing aerosols $\omega_0 = 0$. Or in other word it is the ratio of the scattering coefficient to the extinction coefficient, measuring the relative importance of scattering and absorption.

$$\omega_0 = \frac{\beta_{sca}}{\beta_{ext}}$$

(D) Phase Function: The phase function represents the angular distribution of the scattered energy. It is the ratio of the scattered intensity at a specific direction to the integral of the scattered intensity at all directions. The phase function depends on the orientation of the particle with respect to the direction of the incident radiation

and on the particle characteristics. In principle, given the size distribution of an assumed spherical aerosol, the phase function can be calculated from Mie theory [12].

(E) Asymmetry Factor: This is defined as the average cosine of the scattering angles for scattered radiation. It is denoted by $g = \langle \cos(\theta) \rangle$ and it can have a value between -1 and +1. For particles with isotropic scattering properties $g=0$, when scattering is entirely in the forward direction $g=+1$ and when scattering is entirely in the backward directions $g=-1$.

$$g = \langle \cos(\theta) \rangle$$

(F) Complex Refractive Index: The complex refractive index is given as $m = m_r + im_i$ where, m_r is the real part which represents scattering and the imaginary part m_i represents absorption and both are functions of wavelength and also depend on the chemical composition. Soot or elemental carbon has the largest value of m_i among the atmospheric aerosols and sulfate has very low values of m_i .

Chapter 2

Light Scattering by small spherical particles

In this chapter, we discuss Mie scattering, the far field solution of Mie for light scattering by spherical particles and also discuss to solve field equations for scattered light by spherical particles.

2.1 Mie Scattering

Mie scattering is one of the kinds of atmospheric scattering in which the wavelength of the incident radiation is not changed. It applies to spheres when the sphere is isotropic, and homogeneous. This is the simplest type of scattering since two of the scattering coefficients must identically equal to zero due to symmetry. The five Maxwell equations used to describe effects of the electromagnetic on material objects are:

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

$$\nabla \times E = -\frac{1}{c} \frac{\partial B}{\partial t}$$

$$\nabla \cdot D = 4\pi\rho$$

$$\nabla \cdot B = 0$$

For the case that there are no charge ($\rho = 0$) and current density ($J = 0$), and to the medium which is homogeneous so that ϵ and μ are constants. Thus, Maxwell equations reduces to [10-13]

$$\nabla \times H = \frac{\epsilon}{c} \frac{\partial E}{\partial t} \quad (2.1.1)$$

$$\nabla \times E = \frac{-\mu}{c} \frac{\partial H}{\partial t} \quad (2.1.2)$$

$$\nabla \cdot H = 0 \quad (2.1.3)$$

$$\nabla \cdot E = 0 \quad (2.1.4)$$

Equations (2.1.1) and (2.1.4) will be used to derive the electromagnetic wave equation. Here the Gaussian (cgs) system of units is used.

2.2 The Electromagnetic wave equation and its solution

We consider a plane electromagnetic wave in a periodic field with a circular frequency ω so that we may write

$$E \rightarrow E e^{i\omega t} \quad (2.2.1)$$

$$H \rightarrow H e^{i\omega t} \quad (2.2.2)$$

The curl operations of Eqs.(2.2.2) and (2.2.1) become

$$\nabla \times H = ikm^2 E \quad (2.2.3)$$

$$\nabla \times E = -ikH \quad (2.2.4)$$

where $k = \frac{2\pi}{\lambda}$ is the wave number denoting the propagation constant in vacuum, λ is the wave length in vacuum, $m = \sqrt{\epsilon}$ is the complex refractive index of the medium at the frequency ω , and the permeability $\mu \approx 1$ for air. We now perform the curl operation on Eq.(2.2.4) and Eq.(2.2.3) to obtain

$$\nabla^2 E = -k^2 m^2 E \quad (2.2.5)$$

$$\nabla^2 H = -k^2 m^2 H \quad (2.2.6)$$

Equation (2.2.5) and (2.2.6) indicate that the electric vector and magnetic induction in a homogeneous medium satisfy the vector wave equation

$$\nabla^2 A + k^2 m^2 A = 0 \quad (2.2.7)$$

where A may be either E or H. Defining the scalar function ψ which satisfies the scalar wave equation

$$\nabla^2 \psi + k^2 m^2 \psi = 0 \quad (2.2.8)$$

The vectors M_ψ and N_ψ in spherical coordinates (r, θ, ϕ) are given by

$$M_\psi = \nabla \times [a_r(r\psi)] = a_\theta \frac{1}{r \sin \theta} \frac{\partial(r\phi)}{\partial \psi} - a_\phi \frac{1}{r} \frac{\partial(r\phi)}{\partial \theta} \quad (2.2.9)$$

$$mkN_\psi = \nabla \times M_\psi = a_r \left[\frac{\partial^2(r\psi)}{\partial r^2} + m^2 k^2(r\psi) \right] + a_\theta \frac{1}{r} \frac{\partial^2(r\psi)}{\partial r \partial \theta} + a_\phi \frac{1}{r \sin \theta} \frac{\partial^2(r\psi)}{\partial r \partial \theta} \quad (2.2.10)$$

Assuming that u and v are two independent solutions of the scalar wave equation defined in Eq.(2.2.8), then the electric and magnetic field vectors expressed by

$$E = M_v + N_u \quad (2.2.11)$$

$$H = m(-M_u + iN_v) \quad (2.2.12)$$

Employing Eqs.(2.2.11) and (2.2.12), E and H can be written explicitly as [8,9]

$$\begin{aligned} E &= a_r \frac{i}{mk} \left[\frac{\partial^2(ru)}{\partial r^2} + m^2 k^2(ru) \right] \\ &+ a_\theta \left[\frac{1}{r \sin \theta} \frac{\partial(rv)}{\partial \phi} + \frac{i}{mkr} \frac{\partial^2(ru)}{\partial r \partial \theta} \right] \\ &+ a_\phi \left[\frac{-1}{r} \frac{\partial(rv)}{\partial \theta} + \frac{1}{mkr \sin \theta} \frac{\partial^2(ru)}{\partial r \partial \phi} \right] \end{aligned} \quad (2.2.13)$$

$$\begin{aligned} H &= a_r \frac{i}{k} \left[\frac{\partial^2(rv)}{\partial r^2} + m^2 k^2(rv) \right] \\ &+ a_\theta \left[\frac{-m}{r \sin \theta} \frac{\partial(ru)}{\partial \phi} + \frac{i}{kr} \frac{\partial^2(rv)}{\partial r \partial \theta} \right] \\ &+ a_\phi \left[\frac{m}{r} \frac{\partial(ru)}{\partial \theta} + \frac{i}{kr \sin \theta} \frac{\partial^2(rv)}{\partial r \partial \phi} \right] \end{aligned} \quad (2.2.14)$$

The scalar wave equation in spherical coordinates is given by

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial \psi}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial \psi}{\partial \theta} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial^2 \psi}{\partial \phi^2} + k^2 m^2 \psi = 0 \quad (2.2.15)$$

This equation is separable by letting

$$\psi(r, \theta, \phi) = R(r) \Theta(\theta) \Phi(\phi) \quad (2.2.16)$$

Which when substituted into (2.2.15) yield the three separated equations:

$$\frac{1}{\Phi} \frac{d^2 \Phi}{d\phi^2} = \text{constant} = -l^2 \quad (2.2.17)$$

$$\frac{1}{R} \frac{d}{dr} \left(r^2 \frac{dR}{dr} \right) + k^2 m^2 r^2 = \text{constant} = n(n+1) \quad (2.2.18)$$

$$\frac{1}{\sin \theta} \frac{1}{\Theta} \frac{d}{d\theta} \left(\sin \theta \frac{d\Theta}{d\theta} \right) - \frac{l^2}{\sin^2 \theta} = \text{constant} = -n(n+1) \quad (2.2.19)$$

The single value solution for Eq.(2.2.17) is simply

$$\Phi = a_l \cos l\phi + b_l \sin l\phi = 0 \quad (2.2.20)$$

Where a_l and b_l are arbitrary constants. Eq.(2.2.19) is the well known equation for spherical harmonics. For convenience we introduce a new variable $\mu = \cos \theta$ so that

$$\frac{d}{d\mu} \left[(1 - \mu^2) \frac{d\Theta}{d\mu} \right] + \left[n(n+1) - \frac{l^2}{1 - \mu^2} \right] \Theta = 0 \quad (2.2.21)$$

The solution of Eq.(2.2.19) can be expressed by the associated Legendre polynomials (spherical harmonics of the first kind) in the form [13]

$$\Theta = P_n^1(\mu) = P_n^1(\cos \theta) \quad (2.2.22)$$

If we introduce the dimensionless variable $\rho = kmr$ and define the function

$$R = \left(\frac{1}{\sqrt{\rho}}\right)Z(\rho)$$

$$\frac{d^2 Z}{d\rho^2} + \frac{1}{\rho} + \left[1 - \frac{(n + \frac{1}{2})^2}{\rho}\right]Z = 0 \quad (2.2.23)$$

The linearly independent solutions to Eq. (2.2.18) is then

$$R = \frac{1}{\sqrt{kmr}} Z_{n+\frac{1}{2}}(kmr) \quad (2.2.24)$$

Combining Eqs.(2.2.20), (2.2.22), and (2.2.24), the scalar wave functions at all points on the surface of a sphere are given by

$$\psi(r, \theta, \phi) = \frac{1}{\sqrt{kmr}} Z_{n+\frac{1}{2}}(kmr) P_n^1(\cos \theta) (a_l \cos l\phi + b_l \sin l\phi) \quad (2.2.25)$$

Each cylindrical function denoted in Eq.(2.2.24) may be expressed as a linear combination of two cylindrical functions of standard type, e.g., the Bessel functions $J_{n+\frac{1}{2}}$ and the Neumann functions $N_{n+\frac{1}{2}}(\rho)$:

$$\psi_n(\rho) = \sqrt{\frac{\pi\rho}{2}} J_{n+\frac{1}{2}}, \quad \chi_n(\rho) = -\sqrt{\frac{\pi\rho}{2}} N_{n+\frac{1}{2}} \quad (2.2.26)$$

The functions $\psi_n(\rho)$ are regular in every finite domain of the ρ plane including the origin, whereas the functions χ_n have singularities at the origin $\rho = 0$ where they become infinite. Therefore we use ψ_n but not χ_n to represent the wave inside the sphere. On utilizing the definitions in Eq.(2.2.26), Eq.(2.2.24) can be rewritten in the form

$$rR = c_n \psi_n(kmr) + d_n \chi_n(kmr) \quad (2.2.27)$$

where c_n and d_n are arbitrary constants. Eq(2.2.27) now represents the general solution of Eq.(2.2.18). It follows the general solution of the scalar wave Eq.(2.2.15) then can be expressed by

$$r\psi(r, \theta, \phi) = \sum_{n=0}^{\infty} \sum_{l=-n}^n P_n^1(\cos \theta) [c_n \psi_n(kmr) + d_n \chi_n(kmr)] \times (a_l \cos l\phi + b_l \sin l\phi) \quad (2.2.28)$$

More over, when $c_n = 1$; and $d_n = i$, we note that

$$\psi_n(\rho) + i\chi_n(\rho) = \sqrt{\frac{\pi\rho}{2}} H_{n+\frac{1}{2}}^2 = \xi_n(\rho) \quad (2.2.29)$$

where $H_{n+\frac{1}{2}}$ is the half integral order Hankel function of the second kind. It has the property of vanishing at infinity in the complex plane and is suitable for the representation of the scattered wave.

2.3 Formal Scattering Solution

Here we discuss the scattering of a plane wave by a homogeneous sphere. For simplicity, we assume that outside the medium is vacuum ($m=1$), that the material of the sphere has an index of refraction m , and the incident radiation is linearly polarized. We select the origin of a rectangular system of coordinates at the center of the sphere, with the positive Z axis along the direction of propagation of the incident wave. If the amplitude of the incident wave is normalized to unity, the incident electric and magnetic field vectors are

$$E^i = a_x e^{-ikz}, \quad H^i = a_y e^{-ikz} \quad (2.3.1)$$

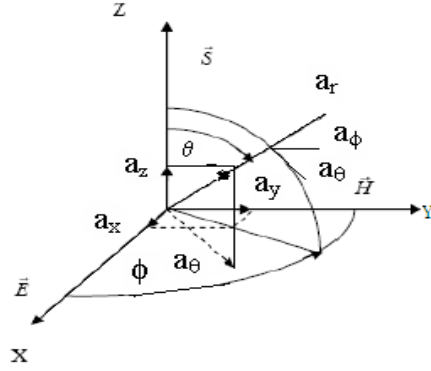


Figure 2.1: Transformation of rectangular to spherical coordinates. $\vec{S}$ is the poynting vector, and $\vec{a}$ is an arbitrary unit vector.

where a_x and a_y are unit vectors along the X and Y axes respectively. The components of any vector say, say $\vec{a}$, in the cartesian coordinate system may be transformed to the spherical polar coordinates (r, θ, ϕ) defined by

$$X = r \sin \theta \cos \phi, \quad Y = r \sin \theta \sin \phi, \quad Z = r \cos \theta \quad (2.3.2)$$

$$\begin{aligned} a_r &= a_x \sin \theta \cos \phi + a_y \sin \theta \sin \phi + a_z \cos \theta, \\ a_\theta &= a_x \cos \theta \cos \phi + a_y \cos \theta \sin \phi - a_z \sin \theta, \\ a_\phi &= -a_x \sin \phi + a_y \cos \phi \end{aligned} \quad (2.3.3)$$

Thus, the electric and magnetic field vectors of the incident wave are

$$\begin{aligned} E_r^i &= e^{-ikr \cos \theta} \sin \theta \cos \phi \\ E_\theta^i &= e^{-ikr \cos \theta} \cos \theta \cos \phi \\ E_\phi^i &= e^{-ikr \cos \theta} \sin \phi \end{aligned} \quad (2.3.4)$$

$$\begin{aligned} H_r^i &= e^{-ikr \cos \theta} \sin \theta \sin \phi, \\ H_\theta^i &= e^{-ikr \cos \theta} \cos \theta \sin \phi, \\ H_\phi^i &= e^{-ikr \cos \theta} \cos \phi \end{aligned} \quad (2.3.5)$$

The first factor on the right-hand side of the above equation may be expressed in the following differentiable series of Legendre polynomials [13]

$$e^{-ikr \cos \theta} \cos \phi = \sum_{n=0}^{\infty} (-i)^n (2n+1) \frac{\psi_n(kr)}{kr} P_n(\cos \theta) \quad (2.3.6)$$

where ψ_n is defined in Eq.(2.2.26). Using the mathematical identities below

$$e^{-ikr \cos \theta} \sin \theta = \frac{1}{ikr} \frac{\partial}{\partial \theta} (e^{-ikr \cos \theta}) \quad (2.3.7)$$

$$\frac{\partial}{\partial \theta} P_n(\cos \theta) = -P_n^1(\cos \theta), \quad P_0^1(\cos \theta) = 0 \quad (2.3.8)$$

Equation (2.3.8) relates the Legendre polynomial P_n with the associated Legendre polynomial P_n^1 . To determine the potential u and v , only one of the components in Eq.(2.2.13) is needed. The first of them is ($m=1$)

$$E_r^i = e^{-ikr \cos \theta} \sin \theta \cos \phi = \frac{i}{k} \left[\frac{\partial^2 (ru^i)}{\partial r^2} + k^2 (ru^i) \right] \quad (2.3.9)$$

Using Eqs.(2.3.7)-(2.3.9), we have

$$e^{ikr \cos \theta} \sin \theta \cos \phi = \frac{1}{(kr)^2} \sum_{n=1}^{\infty} (-i)^{n-1} (2n+1) \psi_n(kr) P_n(\cos \theta) \cos \phi \quad (2.3.10)$$

Taking the trial solution of Eq.(2.3.9) in a series of similar form

$$ru^i = \frac{1}{k} \sum_{n=1}^{\infty} \alpha_n \psi_n(kr) P_n^1(\cos \theta) \cos \phi, \quad (2.3.11)$$

Upon substituting Eqs.(2.3.11) and (2.3.10) into Eq.(2.3.9) and comparing coefficients, we obtain

$$\alpha_n [k^2 \psi_n(kr) + \frac{\partial^2 \psi_n(kr)}{\partial r^2}] = (-i)^n (2n+1) \frac{\psi_n(kr)}{r^2} \quad (2.3.12)$$

In Eq.(2.2.27), since $\chi_n(kr)$ become infinite at the origin through which the incident wave must pass, we may let $c_n = 1$ and $d_n = 0$. Hence

$$\psi_n(kr) = rR \quad (2.3.13)$$

Comparing Eq.(2.3.12) with (2.3.10), we find

$$\alpha_n = (-i)^n \frac{2n+1}{n(n+1)} \quad (2.3.14)$$

Utilizing the similar procedures, v^i can be derived from Eq.(2.2.14). Thus, for incident waves outside the sphere, we have

$$\begin{aligned} ru^i &= \frac{1}{k} \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} \psi_n(kr) P_n^1(\cos \theta) \cos \phi \\ rv^i &= \frac{1}{k} \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} \psi_n(kr) P_n^1 \cos \theta \sin \phi \end{aligned} \quad (2.3.15)$$

For internal waves, because the function $\chi_n(kmr)$ becomes infinite at the origin, only the function $\psi_n(kr)$ may be used. Thus for internal waves we have

$$\begin{aligned} ru^t &= \frac{1}{mk} \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} c_n \psi_n(kr) P_n^1(\cos \theta) \cos \phi, \\ rv^t &= \frac{1}{mk} \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} d_n \psi_n(kr) P_n^1(\cos \theta) \sin \phi \end{aligned} \quad (2.3.16)$$

The Hankel functions expressed in Eq.(2.2.29) will show the scattered properties which vanish at infinity. Therefore, for scattered waves we have

$$\begin{aligned} ru^s &= \frac{-1}{k} \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} a_n \xi_n(kr) P_n^1(\cos \theta) \cos \phi, \\ rv^s &= \frac{-i}{k} \sum_{n=1}^{\infty} (-i)^n \frac{2n+1}{n(n+1)} b_n \xi_n(kr) P_n^1(\cos \theta) \sin \phi, \end{aligned} \quad (2.3.17)$$

The coefficients a_n , b_n , c_n , and d_n have to be determined from the boundary conditions at the surface of the sphere. The conditions are that the tangential components of E and H be continuous across the sphere surface $r=a$, so we have

$$\begin{aligned} E_{\theta}^i + E_{\theta}^s &= E_{\theta}^t, & H_{\theta}^i + H_{\theta}^s &= H_{\theta}^t \\ E_{\phi}^i + E_{\phi}^s &= E_{\phi}^t, & H_{\phi}^i + H_{\phi}^s &= H_{\phi}^t \end{aligned} \quad (2.3.18)$$

Equation (2.3.18) implies that these four expressions have to be continuous at $r=a$.

Consequently,

$$\begin{aligned} \frac{\partial}{\partial r} [r(u^i + u^s)] &= \frac{1}{m} \frac{\partial}{\partial r} (ru^t), & u^i + u^s &= mu^t \\ \frac{\partial}{\partial r} [r(v^i + v^s)] &= \frac{\partial}{\partial r} (rv^t), & v^i + v^s &= v^t \end{aligned} \quad (2.3.19)$$

From these equations, we obtain

$$\begin{aligned}
m[\psi'_n(ka) - a_n \xi'_n(ka)] &= c_n \psi'_n(kma) \\
[\psi'_n(ka) - b_n \xi'_n(ka)] &= d_n \psi'_n(kma) \\
[\psi_n(ka) - a_n \xi_n(ka)] &= c_n \psi_n(kma) \\
m[\psi_n(ka) - b_n \xi_n(ka)] &= d_n \psi_n(kma)
\end{aligned} \tag{2.3.20}$$

where the prime denotes differentiation with respect to the argument, upon eliminating c_n and d_n , we obtain the coefficients for the scattered waves in the forms

$$\begin{aligned}
a_n &= \frac{\psi'_n(y)\psi_n(x) - m\psi_n(y)\psi'_n(x)}{\psi'_n(y)\xi_n(x) - m\psi_n(y)\xi'_n(x)} \\
b_n &= \frac{m\psi'_n(y)\psi_n(x) - \psi_n(y)\psi'_n(x)}{m\psi'_n(y)\xi_n(x) - \psi_n(y)\xi'_n(x)}
\end{aligned} \tag{2.3.21}$$

where $x=ka$ and $y=mx$, x is called the size parameter.

2.4 The Far field solutions and extinction parameters

We shall now consider the far field at very large distances from the sphere. We note that for all practical applications, all light scattering observations are normally carried out in the far-field zone. In the far field the Hankel functions denoted in Eq.(2.3.2) reduce to the form

$$\xi_n(kr) = i^{n+1} e^{-ikr}, \quad kr \gg 1 \tag{2.4.1}$$

With this simplification Eq.(2.3.17) becomes

$$\begin{aligned} ru^s &\approx -\frac{-ie^{-ikr} \cos \phi}{k} \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} a_n P_n^1(\cos \theta) \\ rv^s &\approx -\frac{-ie^{-ikr} \sin \phi}{k} \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} b_n P_n^1(\cos \theta) \end{aligned} \quad (2.4.2)$$

The three components of the electric and magnetic field vectors in Eq.(2.2.13) and (2.2.14) are given by

$$\begin{aligned} E_r^s = H_r^s &\approx 0 \\ E_\theta^s = H_\phi^s &\approx \frac{-ie^{-ikr} \cos \phi}{kr} \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n \frac{dP_n^1(\cos \theta)}{d\theta} + b_n \frac{P_n^1(\cos \theta)}{\sin \theta} \right] \\ -E_\phi^s = H_\theta^s &\approx \frac{-ie^{-ikr} \sin \phi}{kr} \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n \frac{P_n^1(\cos \theta)}{\sin \theta} + b_n \frac{dP_n^1(\cos \theta)}{d\theta} \right] \end{aligned} \quad (2.4.3)$$

We find that the radial components E_r^s and H_r^s may be neglected in the far-field zone.

Defining two scattering functions of the forms

$$\begin{aligned} S_1(\theta) &= \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n \pi_n(\cos \theta) + b_n \tau_n(\cos \theta)] \\ S_2(\theta) &= \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [b_n \pi_n(\cos \theta) + a_n \tau_n(\cos \theta)] \end{aligned} \quad (2.4.4)$$

where $\pi_n(\cos \theta) = \frac{1}{\sin \theta} P_n^1(\cos \theta)$, $\tau_n(\cos \theta) = \frac{d}{d\theta} P_n^1(\cos \theta)$. Thus, we may write

$$\begin{aligned} E_\theta^s &= \frac{-i}{kr} e^{-ikr} \cos \phi S_2(\theta) \\ -E_\phi^s &= \frac{-i}{kr} e^{-ikr} \sin \phi S_1(\theta) \end{aligned} \quad (2.4.5)$$

These fields represent an outgoing spherical wave with amplitude and state of polarization as functions of the scattering angle θ . By defining the two polarization states, parallel $E_{\parallel}$ and perpendicular $E_{\perp}$. Eq.(2.4.5) becomes

$$\begin{bmatrix} E_{\parallel}^s \\ E_{\perp}^s \end{bmatrix} = \frac{e^{-ikr+ikz}}{ikr} \begin{bmatrix} S_2(\theta) & 0 \\ 0 & S_1(\theta) \end{bmatrix} \begin{bmatrix} E_{\parallel}^i \\ E_{\perp}^i \end{bmatrix} \quad (2.4.6)$$

The scattered intensity components in the far-field zone can now written in terms of the incident intensity components in the form

$$I_{\parallel}^s = I_{\parallel}^i \frac{i_2}{k^2 r^2}, \quad I_{\perp}^s = I_{\perp}^i \frac{i_1}{k^2 r^2} \quad (2.4.7)$$

where $i_1(\theta) = |S_1(\theta)|^2$, $i_2(\theta) = |S_2(\theta)|^2$ and they are called the intensity functions for the perpendicular and parallel components, respectively. In the far-field zone, we would like to evaluate the reduction of the incident energy due to absorption and scattering of light by a sphere. For this purpose we consider incident light polarized linearly in the perpendicular direction. From Eq.(2.4.6) the scattered electric field is given by

$$E_{\perp}^s = \frac{e^{-ikr+ikz}}{ikr} S_1(\theta) E_{\perp}^i \quad (2.4.8)$$

Consider a point (x,y,z) in the forward direction, i.e., $\theta = 0$. In the far field since $x(y) \ll z$, we have in the forward direction

$$r = (x^2 + y^2 + z^2)^{\frac{1}{2}} \approx z + \frac{x^2 + y^2}{2z} \quad (2.4.9)$$

Upon superimposing the incident and scattered electric fields in the forward direction. we obtain

$$E_{\perp}^i + E_{\perp}^s \approx E_{\perp}^i \left\{ 1 + \frac{S_1(0)}{ikz} e^{-ik(\frac{x^2+y^2}{2z})} \right\} \quad (2.4.10)$$

The far field field combined flux density in the forward directions then is proportional to

$$|E_{\perp}^i + E_{\perp}^s|^2 \approx |E_{\perp}^i|^2 \left\{ 1 + \frac{2}{kz} \text{Re} \left[\frac{S_1(0)}{i} e^{-ik(\frac{x^2+y^2}{2z})} \right] \right\} \quad (2.4.11)$$

Integrating the combined flux density over the cross section area of a sphere whose radius is $r=a$, we obtain the total power combined:

$$\frac{1}{|E_{\perp}^i|^2} \int \int |E_{\perp}^i + E_{\perp}^s|^2 dx dy = \pi a^2 - \sigma_e \quad (2.4.12)$$

where the first term on the right-hand side of Eq.(2.4.12) represents the cross section of area of the sphere. The physical interpretation of the second term σ_e is that the total light received in the forward direction is reduced by the presence of the sphere, and the amount of the reduction is as if an area σ_e of the object had been covered up. The double integral over $dx dy$ by which σ_e is defined contains two Fresnel integrals, and if the limits are assumed to extend to ∞ , we get

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{-ik(\frac{x^2+y^2}{2z})} dx dy = \frac{2\pi z}{ik} \quad (2.4.13)$$

Thus, the extinction cross section is

$$\sigma_e = \frac{4\pi}{k^2} \text{Re}[S(0)] \quad (2.4.14)$$

The two amplitude functions in the forward directions reduce to $S_1(0) = S_2(0) = S(0) = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1)(a_n + b_n)$. The fact that there is only one $S(0)$ is because of the symmetry of the forward scattering in which the extinction is independent of the state of polarization of the incident light. It should be noted that Eq.(2.4.14) is valid when the sphere is isotropic and homogeneous. Define the extinction efficiency for a sphere with a radius of $r=a$ is

$$Q_e = \frac{\sigma_e}{\pi a^2} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \text{Re}[a_n + b_n] \quad (2.4.15)$$

where $x=ka$ is the size parameter which is discussed in chapter one. The flux density of the scattered light in an arbitrary direction is given by

$$F(\theta, \phi) = \frac{F_0}{k^2 r^2} [i_2(\theta) \cos^2 \phi + i_1(\theta) \sin^2 \phi] \quad (2.4.16)$$

with $F_0=1$ (unit amplitude). The total flux (or power) of the scattered light is therefore

$$f = \int_0^{2\pi} \int_0^\pi F(\theta, \phi) r^2 \sin \theta d\theta d\phi \quad (2.4.17)$$

where $\sin \theta d\theta d\phi$ is the differential solid angle $d\Omega$, and $r^2 d\Omega$ depends the differential area. Hence, the scattering cross section may be defined as

$$\sigma_s = \frac{f}{F_0} = \frac{\pi}{k^2} \int_0^\pi [i_1(\theta) + i_2(\theta)] \sin \theta d\theta \quad (2.4.18)$$

In a similar way the in extinction case, we define the scattering efficiency for a sphere

$$Q_s = \frac{\sigma_s}{\pi a^2} = \frac{1}{x^2} \int_0^\pi [i_1(\theta) + i_2(\theta)] \sin \theta d\theta \quad (2.4.19)$$

Using the orthogonal and recurrence properties of the associated Legendre polynomials [11-13]:

$$\int_0^\pi \left(\frac{dP_n^1}{d\theta} \frac{dP_m^1}{d\theta} + \frac{1}{\sin^2 \theta} P_n^1 P_m^1 \right) \sin \theta d\theta = \begin{cases} 0, & \text{if } n \neq m ; \\ \frac{2n(n+1)(n+1)!}{(2n+1)(n-1)!}, & \text{if } n=m. \end{cases} \quad (2.4.20)$$

and

$$\int_0^\pi \left(\frac{P_n^1}{\sin \theta} \frac{dP_m^1}{d\theta} + \frac{P_m^1}{\sin \theta} \frac{dP_n^1}{d\theta} \right) \sin \theta d\theta = [P_n^1(\theta) P_m^1(\theta)]_0^\pi = 0 \quad (2.4.21)$$

The scattering efficiency can be evaluated with the help of these two equations yield

$$Q_s = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) (|a_n|^2 + |b_n|^2) \quad (2.4.22)$$

Finally, the absorption cross section and efficiency of a sphere can be calculated from

$$\sigma_a = \sigma_e - \sigma_s, \quad Q_a = Q_e - Q_s$$

$$\sigma_a = \frac{\pi}{k^2} \left| \sum_{n=1}^{\infty} (2n+1) (-1)^2 (a_n - b_n) \right|^2 \quad (2.4.23)$$

$$Q_a = \frac{1}{x^2} \left| \sum_{n=1}^{\infty} (2n+1) (-1)^n (a_n - b_n) \right|^2 \quad (2.4.24)$$

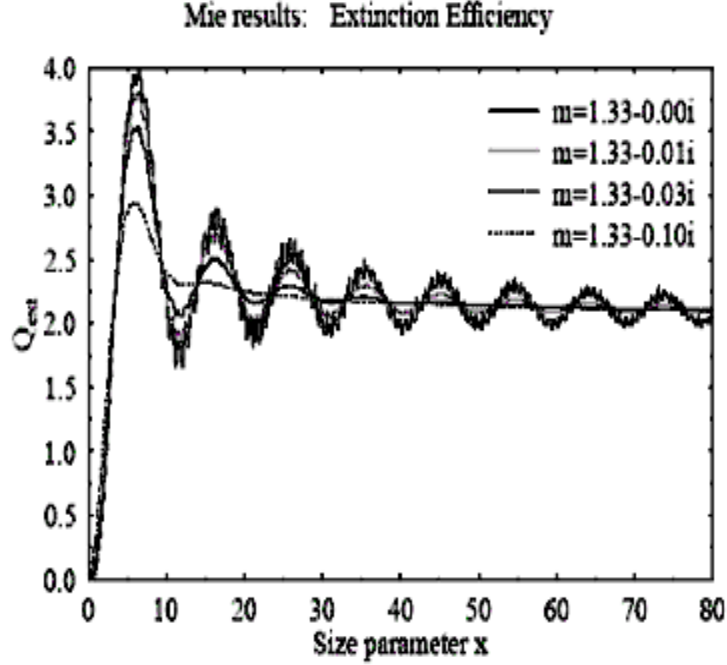


Figure 2.2: Mie results for different values of the imaginary refractive index.

Figure.(2.2). shows the extinction efficiency factor Q_{ext} as a function of the size parameter x for a real index of refraction 1.33 with several values of the imaginary part. For $m_i=0$, i.e., a perfect reflector, there is no absorption so that $Q_{ext} = Q_s$. Q_{ext} in this case shows a series of major maxima and minima and ripples. The major maxima and minima are due to interference of light diffracted and transmitted by the sphere, whereas the ripple arises from edge rays that are grazing and traveling the sphere, spewing off energy in all directions. Both the ripples and the major maxima

and minima damp out as absorption within the particle increases.

Chapter 3

Retrieval theory for deriving aerosol size parameters from extinction data

Retrieving aerosol parameters is important to determine the various properties of aerosols e.g dynamical, optical, electrical and chemical properties. For example retrieving the aerosol size is important to evaluate the optical properties of aerosols.

3.1 The inverse problem

The retrieval theory is developed to solve an inverse problem. There are systems in which we are unable to measure directly the quantity we are interested in e.g. the state vector x representing, aerosol size distribution but are able to measure some other quantity somehow related to or resulting from it, e.g. measurement vector y representing optical depth at various wavelength. From this measurement y we

wish to find out some thing about the state x the system is in. Given $y=F(x)$ for some function F known as the forward function, approximated by the Forward model (FM) $f(x)$, solving the inverse problem means being able to find the inverse function $x = f^{-1}(y)$ and so obtain state information from measurement y . The premise behind performing an aerosol retrieval is that aerosol in the atmosphere will affect the light received at the satellite through a combination of absorption and scattering. Aerosol radiative properties depend on factors such as aerosol particle size, shape, chemical composition and vertical profile. Therefore a forward model must take such factors into account. The use of satellite data or any other remotely measured data by sensors on ground, aircraft, spacecraft, research vessel etc, in this way is known as remote sensing (or remote sounding), as the measurements are taken at distance.

3.1.1 Retrieval formalism

The inverse problem for forward model is an ill-posed problem which fail to satisfy the following conditions: uniqueness, stability, and direct inverse of the solution [14-16]. A sample of atmospheric aerosol can be described by its number density, size and shape distribution, vertical profile and chemical make-up (which determines refractive index). Assuming the particles are spherical allows computation of radiative properties from Mie theory. Aerosol optical depth is a complex function of particle characteristics, such as size distribution, shape, and chemical composition. Aerosol optical depth is defined as

$$\tau(\lambda) = \int_0^\infty \beta_{ext}(z, \lambda) dz = \int_0^\infty (\beta_{sca} + \beta_{abs}) dz \quad (3.1.1)$$

The total extinction coefficient β_{ext} , is defined as the sum of the extinction due to absorption β_{abs} and scattering β_{sca} . Radiative characteristics are described by the

vertical profiles of β_{abs} and β_{sca} along with the scattering phase functions $P(\theta)$ which determines the spatial distribution of the scattered radiation.

3.1.2 Retrieval of aerosol size distributions from optical thickness measurements

Optical thickness spectra carry information about the aerosol size distribution. Assuming that aerosols are homogeneous spheres, the equation that relates these two aerosol properties is

$$\tau_m(\lambda) = \int_0^\pi \pi r^2 Q_{ext}(r, \lambda, m) n(r) dr \quad (3.1.2)$$

where $\tau_m(\lambda)$ is the aerosol optical thickness, $n(r)$ is the unknown columnar aerosol size distribution, m is the complex refractive index of the aerosol particles, λ is the wavelength of the incident radiation, $Q_{ext}(r, \lambda, m)$ is the Mie extinction efficiency. The integral in Eq.(3.1.2) corresponds to the Fredholm integral equation of the first kind, which is difficult to solve. Since $n(r)$ can not be written analytically, a numerical approach is usually followed to separate $n(r)$ into two parts, $n(r)=h(r)f(r)$, where $h(r)$ is rapidly varying function of r , while $f(r)$ is more slowly varying. In this work the kind of size distribution used is power law distribution for which $n(r) = cr^{-3}$. Substituting $n(r)$ into Eq.(3.1.2) we get

$$\begin{aligned} \tau_m(\lambda) &= \int_{r_a}^{r_b} \pi r^2 Q_{ext}(r, \lambda, m) h(r) f(r) dr \\ &= \sum_{j=1}^q f(r_j) \int_{r_j}^{r_{j+1}} \pi r^2 Q_{ext}(r, \lambda, m) h(r) dr \end{aligned} \quad (3.1.3)$$

where the limits of integration have to be finite with $r_1 = r_a$ and $r_{q+1} = r_b$. If $f(r)$ is assumed constant within each coarse interval the quadrature error will be less. In

this case a system of linear equations results which may be written as

$$\tau = Af + \varepsilon \quad (3.1.4)$$

where ε is unknown error vector whose elements ε_i represents the deviation between the measurement τ_i and theory $(\sum_j A_{ij}f_j)$. This deviation arises from quadrature and measurement errors, as well as any uncertainties as to the exact form of the kernel functions [in this case, the kernel function is $\pi r^2 Q_{ext}(r, \lambda, m)$]. Therefore using Eq.(3.1.2) the elements of Eq.(3.1.4) are given by

$$\tau_i = \tau_m(\lambda_i), \quad i = 1, 2, \dots, p$$

$$A_{ij} = \int_{r_j}^{r_{j+1}} \pi r^2 Q_{ext}(r, \lambda_i, m) h(r) dr, \quad j = 1, 2, \dots, q \quad (3.1.5)$$

$$f_j = f(\bar{r}_j), \quad r_j \leq \bar{r}_j \leq r_{j+1}$$

The experimental determination of extinction is possible only at discrete values of λ_i . The problem is solved in a number of iterations. The weighting function $h(r)$ is given by the Power law

$$h(r) = r^{-3} \quad (3.1.6)$$

Therefore the solution is found in the form for a linear problem

$$f = (A^T A)^{-1} A^T \tau \quad (3.1.7)$$

However, quite after the relation between f and optical depth is nonlinear $A^T A$ is not invertible. This requires inclusion of stabilizing term γH so that Eq.(3.1.7) becomes

$$f = (A^T A + \gamma H)^{-1} A^T \tau \quad (3.1.8)$$

Where, γ is the Lagrange multiplier, and H is the elements of the smoothing matrix which is given by

$$\begin{pmatrix} 1 & -2 & 1 & 0 & 0 & 0 \\ -2 & 5 & -4 & 1 & 0 & 0 \\ 1 & -4 & 6 & -4 & 1 & 0 \\ 0 & 1 & -4 & 6 & -4 & 1 \\ 0 & 0 & 1 & -4 & 5 & -2 \\ 0 & 0 & 0 & 1 & -2 & 1 \end{pmatrix} \quad (3.1.9)$$

Eq.(3.1.8) is used to obtain the size distribution array. Since the optical depth is not linearly related with the solution vector f_j , we have to use non linear retrieval procedures as in equation below:

$$\begin{aligned} \delta f &= (A^T A + \gamma H)^{-1} A^T \delta \tau, & \delta \tau &= \tau_{measured} - \tau_{simulated} \\ f_{n+1} - f_n &= (A^T A + \gamma H)^{-1} A^T (\tau_{measured} - \tau_{simulated}) \end{aligned} \quad (3.1.10)$$

Chapter 4

Measurement and simulation of extinction efficiency of spherical aerosol particles

4.1 Simulation of extinction efficiency by spherical particles

Aerosol scattering, absorption, and extinction coefficients are functions of particle size , the complex refractive index of the particles, and the wavelength of the incident radiation. The extinction spectrum $\tau(\lambda)$ of aerosol spherical particles is given by

$$\tau(\lambda) = \int_0^\infty \beta_{ext} n(r) dr \quad (4.1.1)$$

where $\beta_{ext}[\mu m^2]$ is the scattering cross-section kernel model. The kernel functions reflect shape, size and material composition of the particles. Mie scattering cross-section kernel functions have been discussed in great depth in chapter 2.

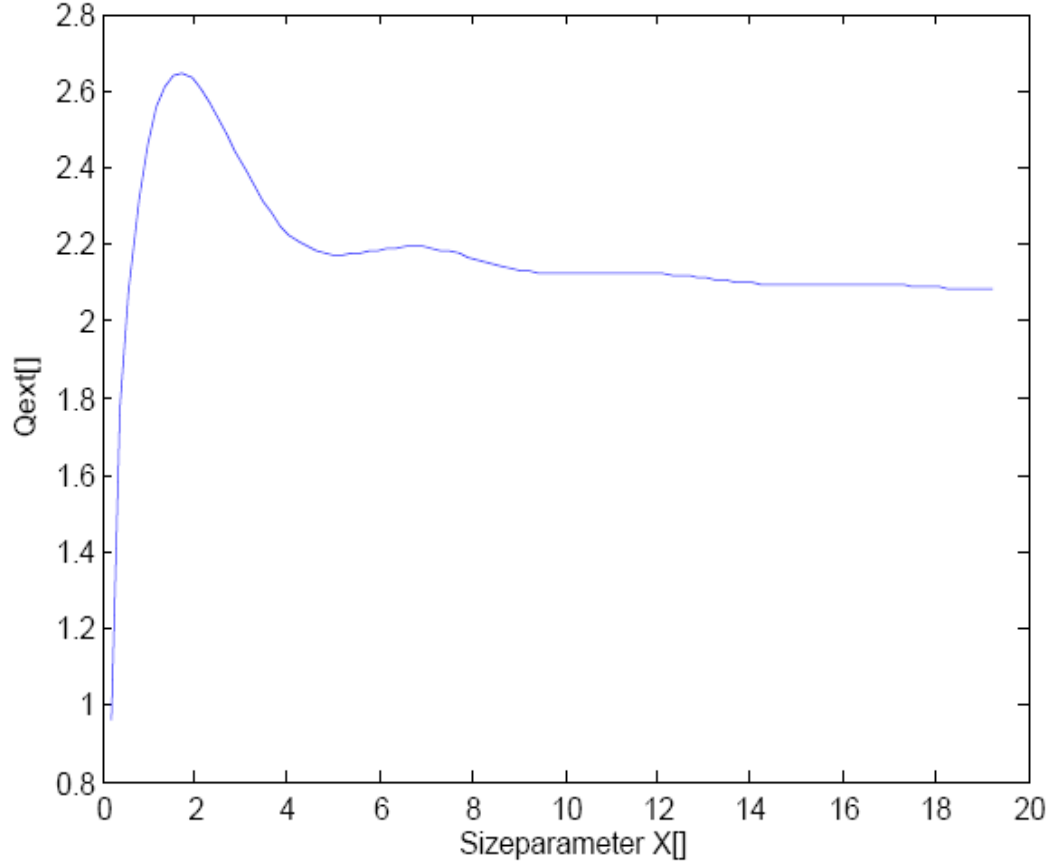


Figure 4.1: The extinction efficiency for water droplets at radius $r=0.5$, $n=1.33-i0.1$ at $\lambda = 0.56$ using the power law distribution

Fig.4.1. Shows the extinction spectrum for water droplets in the region between $0.5 \mu m$ and $5 \mu m$ the spectrum has exhibited the non-absorbing nature of water droplets in this wavelength range. The extinction efficiency becomes maximum when the size parameter approaches two. For larger values of size parameter the extinction curve asymptotically attains a value 2. The ripple structure shown in chapter 2 do not appear in our simulations due to a course grid of size parameter that is used in

the calculations, moreover, ripple structures are not focus of this thesis.

4.2 Optical thickness

The attenuation of electromagnetic radiation in the Earth's atmosphere can be simply described as a reduction of flux density I due to extinction. An incident flux density I_0 of monochromatic radiation is attenuated to

$$I(\lambda) = I_0(\lambda)e^{-\tau(\lambda)M} \quad (4.2.1)$$

where $I(\lambda)$ is the flux density of radiation detected at the ground; I_0 is the extraterrestrial solar radiation, $\tau(\lambda)$ is the atmospheric spectral optical thickness at a given wavelength; and M is the optical air mass. Optical depth is a dimensionless parameter which quantifies the attenuation of the sun's direct beam. A total optical depth of one would attenuate the direct beam by a factor of $e^{-1} = 0.368$.

Total measured optical thickness is defined as

$$\tau(\lambda) = \tau_A + \tau_G + \tau_R \quad (4.2.2)$$

where τ_A is the aerosol optical thickness by scattering and absorption, τ_R is the Rayleigh component of the total optical thickness due to scattering, τ_G is the optical thickness by absorption of atmospheric gases. Molecular optical thickness depends only up on surface pressure and wavelength. Gaseous optical thickness depends on the column amount of the gas multiplied by its absorption cross-section at that wavelength. Gases such as O_3 , NO_2 , and SO_2 absorb sunlight at a certain wavelength region of the spectrum. Therefore, the aerosol optical thickness is found by subtracting the molecular scattering and gaseous absorption from the total measured optical

thickness. For a clear sky the optical thickness due to gaseous absorption can be calculated for each wavelength allowing the aerosol optical depth to be separate.

$$\tau_{\lambda} = \tau_{500} \left(\frac{\lambda}{500} \right)^{-\beta} \quad (4.2.3)$$

where τ_{500} is total aerosol optical thicknes at 500nm; β is measure of the average radius of the aerosol size distribution. Aerosol optical depth product is a measure of how much light air born particles prevent from passing through a column of atmosphere. Aerosols tend to absorb or reflect the incoming sunlight, thus reduce visibility and increase optical depth. Optical depth of less than 0.1 indicates a crystal clear sky with maximum visibility where as the value 4 indicates the presence of aerosol so dense which is difficult to see the sun at mid-day.

4.3 Extinction data from Ground-based network

The Aerosol Robotic Network (AERONET) program is a federated remote sensing network of well-calibrated sun photometers and radiometers. AERONET includes about 200 sites around the world, covering all major tropospheric aerosol regimes [17]. It provides globally distributed near real time observations of aerosol spectral optical thickness and sky radiance as well as derived parameters such as particle size distributions in a manner suitable for integration with satellite data. This network has been developed to provide aerosol information from two kinds of ground-based measurements: spectral data of direct sun radiation extinction (i.e., aerosol optical thickness) and angular distribution of sky radiance. The network imposes standardization of instruments, calibration and processing. The data is centrally archived and available in three processing levels unscreened (L1.0), cloud-screened (L1.5) and

cloud-screened and quality-assured (L2.0). According to Holben et al [17], the accuracy is about ± 0.02 . The data is found on the AERONET project web page (<http://aeronet.gsfc.nasa.gov/data-menu.html>) [3].

4.4 Sensors functionality at AERONET

A sun photometer is an instrument which, when pointed directly at the sun, measures the intensity of sun's light. Sun photometers absorb sunlight energy with the light-emitting Diode (LED) and convert the intensity into a quantified voltage to measure aerosol in the atmosphere. The intensity of sunlight at the top of the earth's atmosphere is constant. While the sun light travels through the atmosphere, though aerosols can dissipate the energy by scattering and absorbing the light. More aerosols in the atmosphere cause more scattering and less energy transmitted to the surface. Therefore, knowing the sunlight's energy at the top of the atmosphere, and the amount of sunlight transmitted to the earth's surface and allows us to determine the amount of scattering, and thus the amount of aerosols. The simulator can be used to practice the aerosol optical thickness. The calibration constant (V_0) for the virtual photometer is 2.117V for both channels. This is also called the extraterrestrial constant because it is the voltage that would be produced by the sun photometer if it were outside of the earth's atmosphere. The decrease in the voltage measured by the sun photometer is the result of light scattered both the aerosol particles and the air molecules that make up the atmosphere. Mathematically, this "total optical thickness" is the sum of the aerosol optical thickness (AOT) and the non-aerosol optical thickness.

The total optical thickness can be from calculated the following equation

$$\textit{Total optical thickness} = \sin(\theta)[\ln V_0 - \ln(V - V_D)]$$

where V_0 is the calibration constant described above, V is the sunlight voltage, V_D is the dark voltage, and θ is the sun angle, once the total optical thickness is calculated, the AOT can be calculated by subtracting the non-aerosol optical thickness from the total optical thickness

$$AOT = \textit{Total optical thickness} - \textit{Nonaerosol optical thickness}$$

The non-aerosol thickness can be read directly from the simulator, or calculated using the following equation

$$\textit{Non-aerosol optical thickness} = a_R\left(\frac{p}{p_0}\right)$$

where a_R is the Rayleigh scattering (non-aerosol optical thickness) at standard sea-level atmospheric pressure (0.138 for the green channel and 0.0058 for the red channel), p is actual atmospheric pressure, and p_0 is standard sea-level atmospheric pressure (1013 millibars).



Figure 4.2: sun photometer Instrument

4.5 Real extinction data

The extinction data obtained from NASA by Brent-Holben in September 2006, for latitudes = 26.208°N , longitudes = 50.609°E , elevation=25m, over location of Bahrain which is situated on the western side of the Persian Gulf and extends between Saudi Arabia and Qatar. The Persian Gulf it self extends from Euphrates River delta to strait of Hormuz. The aerosol in Bahrain (Persian Gulf) has a larger fine mode than observed in Saudi Arabia and Cape Verde. This difference relates to the frequent presence in the Persian Gulf of small particles produced by industrial activity. Using this measured extinction data we have retrieved the aerosol size parameters using the algorithms developed in section 3.1.2. The measurements were made at several different wavelengths but in our case we take only six discrete wavelengths available

on the data of the measurements. These are $0.34\ \mu m$, $0.38\ \mu m$, $0.44\ \mu m$, $0.67\ \mu m$, $0.87\ \mu m$, and $1.02\ \mu m$. We also include the refractive index of aerosols in the range between 1.33 to 1.56 for testing our retrieval algorithm and studying the dependence of aerosol size parameters on refractive index.

4.6 Simulated optical thickness

The Forward model based on Mie theory is used to generate kernel functions for $m=1.33, 1.56, 1.55, 1.53$. The optical thickness is simulated at 6 wavelengths between $0.34\ \mu m$ and $1.02\ \mu m$.

Chapter 5

Results and Discussion

In this thesis, we have simulated the aerosol optical thickness and retrieved aerosol number density from measured extinction data using retrieval algorithms that we have developed.

5.1 Retrieval of aerosol size parameters for power law distribution

As we have already discussed in section 4.2, the aerosol optical thickness are obtained by subtracting the molecular scattering and gaseous absorption from the total measured optical thickness. The aerosol optical depth is highly variable in space and time and it is a complex function of refractive index and particle radius. The power law distribution is used in this thesis to retrieve the aerosol size parameters which is stated in section 3.1.2.

For all inversions, we assumed the complex refractive index of the aerosol particles which is wavelength and size independent and taking the different refractive index in the range between 1.33 and 1.56 for most aerosol types. But the choice of refractive

index and the maximum radius of the distribution is specified by the user because it is the critical determinant of the Mie calculation [12]. There is no absolute rule which determines the radius limits having the most significant contribution to the attenuation measurements. Therefore, we have fixed the wavelength limits to $\pm 0.2 \mu m$ of the particle radius interacting with radiation field.

The aerosol optical thickness measurements made at six different wavelengths ranging between $0.34 \mu m$ and $1.02 \mu m$ have been used in the determination of the size distributions. The final optical thickness determined from a combined use of forward radiative and retrieved aerosol size parameters. As we have discussed in chapter 3, the retrieval procedure allows the particle size parameters to vary until the difference between the measured and the simulated optical thickness is below preset minimum value.

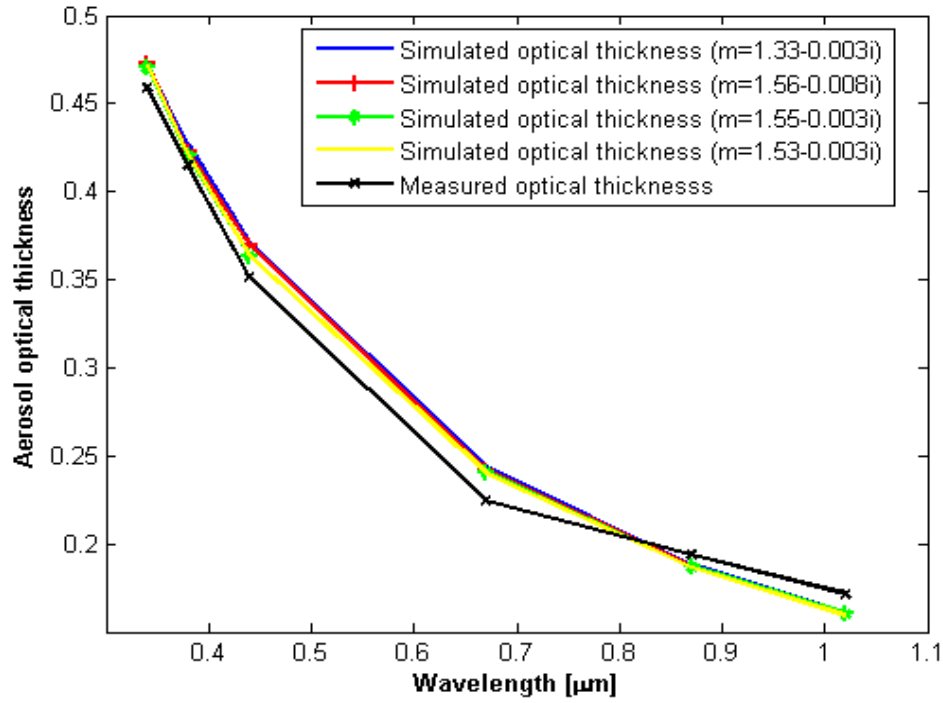


Figure 5.1: Comparison of the measured and simulated optical depth.

The results obtained by using different refractive indices (see Fig.5.1) show that aerosol with real refractive index of 1.53 is the dominant scatterer in the column of air above the site of observation. This refractive index corresponds to dust particles. Further more, as can be seen from Fig.5.1, the aerosol optical thickness varies with wavelength. The highest optical thickness is shorter wavelengths and it decreases towards longer wavelengths. This shows that, in general, particularly at Bahrain small particles dominate. This result agrees with existing frequent presence of small particles in the Persian Gulf produced by industrial activity. And several models suggest that the real part of the complex refractive index of dust is 1.53 for the

visible spectral region [18-20]. This value is usually employed in remote sensing and allows for good consistency of radiative transfer computations with measurements. However, the in situ measured values reported in literature may deviate from 1.53 (due to differences in dust composition and probably also due to differences in the measurement techniques) with a range of about ± 0.05 or more [2]. The maximum aerosol optical thickness is observed at wavelength $0.34 \mu m$ which is in between 0.45 and 0.5 and the lowest is observed at a wavelength of $1.02 \mu m$.

5.2 Effects of refractive index on retrieval of aerosol size parameters

So far, refractive indices are assumed to be accurately known in the size distribution retrieval. In reality, our knowledge of refractive index, however, is uncertain. For aerosols, refractive index is essentially unknown since atmospheric aerosols are mixtures of a variety aerosol types and chemical compositions (e.g., sulfate, nitrate, ammonium, condensed organic species, elemental or black carbon, and mineral dust). Refractive index generally varies with particle radius due to chemical composition of particles of different sizes, this affects the extinction spectrum which is depicted in the Fig.5.1.

Fig 5.2, shows the dependence of columnar size distribution on the types of aerosol in the column of air. The columnar size of dust aerosol contributing to a given optical thickness is smaller than that of water soluble aerosol at the same size. This difference (see Fig.5.2) becomes even more pronounced at smaller particle radius.

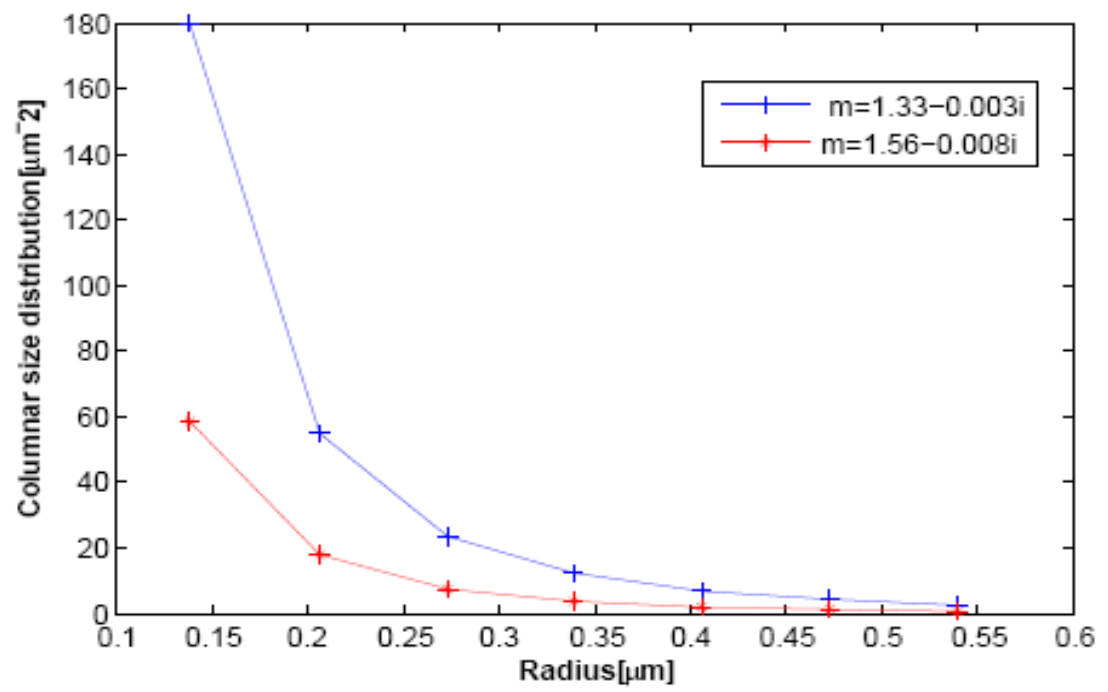


Figure 5.2: Dependence of retrieved size distribution on index of refraction

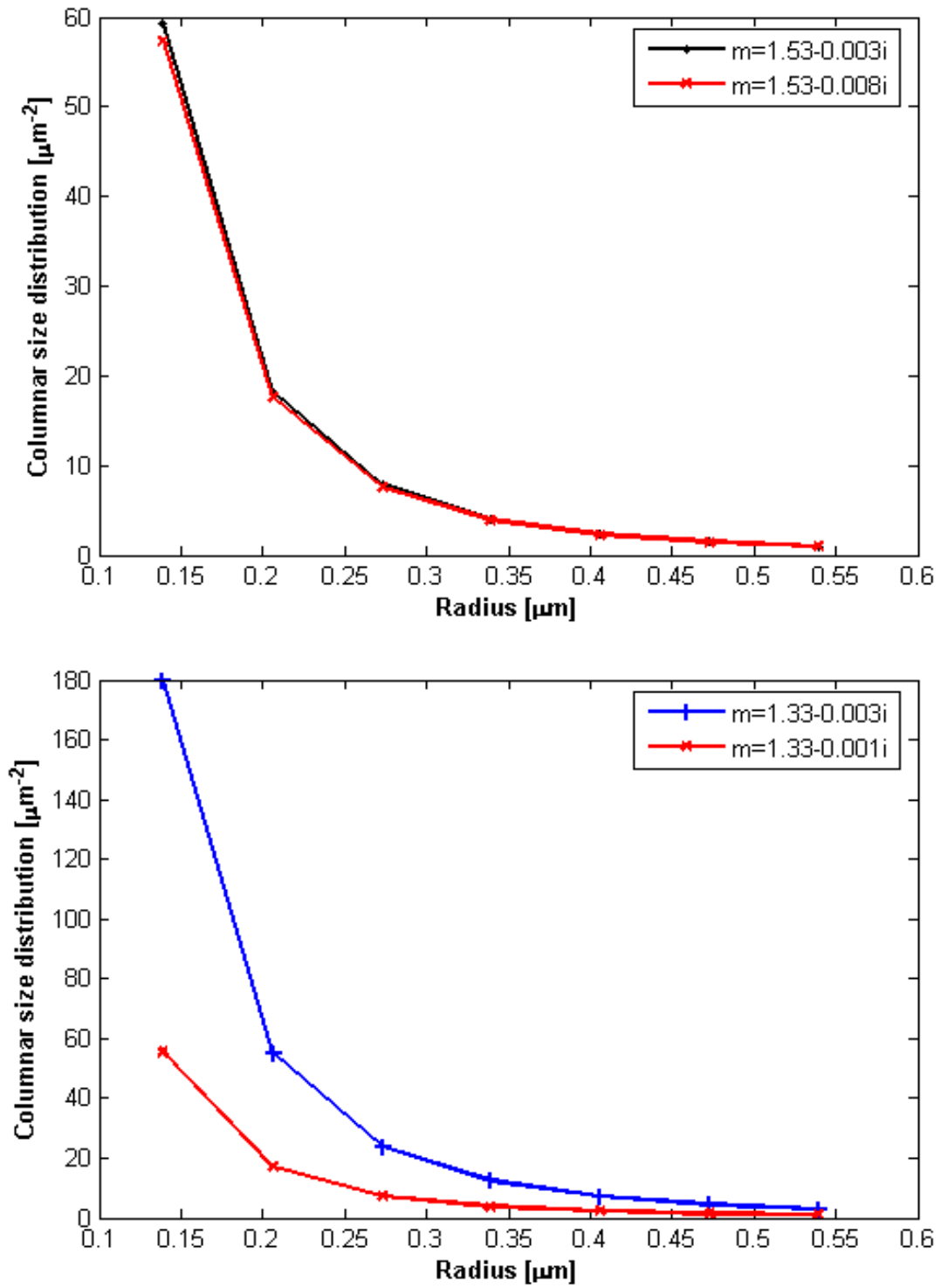


Figure 5.3: The variation of imaginary refractive index and its effect on its retrieved size distribution.

We have highlighted that even with the same aerosol type, the refractive index may vary due to slight change in chemical composition. On account of this reality, we have studied the impact of variations refractive index for both dust and water soluble aerosol shown in Fig.5.3. The results have revealed dust aerosol is weakly sensitive to change in imaginary refractive index. On the contrary, water soluble aerosol have exhibited remarkable variations with imaginary refractive index particularly for small radii particles.

5.3 Phase function of aerosol particles

The flux density of the radiation scattered into an elementary solid angle $d\Omega = \sin \theta d\theta d\phi$ is proportional to $p(\theta, \phi)$, where p is the phase function. The phase function depends on the orientation of the particle with respect to the direction of the incident radiation and on the particle characteristics: the angles θ and ϕ correspond to the direction of the propagation of the scattered radiation for a given particle orientation. The scattering angle θ is simply the angle between the incident beam and the scattered beam ($\theta = 0$ for forward scattering, $\theta = \pi$ for back scattering). The scattering angle ϕ specifies the orientation of the scattering plane relative to the reference plane in the Lab Frame. Within the scattering plane scattering directions are specified by $0 \leq \theta \leq \pi$. The scattering phase function describes the angle dependent scattering of light incident on the particle. The phase function describes the redistribution of radiative energy into space after light-particle interaction. However, the the total energy lost is proportional the cross-section for extinctions β_{ext} . The part proportional to β_{abs} (absorption) is completely lost (a great part is spontaneously released in the form of thermal radiation at different wavelengths) and the part proportional to

$\beta_{ext} - \beta_{abs} = \beta_{sca}$ (scattering) is diffracted (elastic scattering).

The phase function can also be related with the asymmetry parameter. Asymmetry parameter is define as the average cosine of the scattering angle. It is denoted by $g = \langle \cos \theta \rangle$. For a particle that scatters light isotropically (i.e., the same in all directions), g vanishes; g also vanishes if the scattering is symmetric about a scattering angle of 90° . If the particle scatters more light toward the forward direction ($\theta = 0^\circ$), g is positive; g is negative if the scattering is directed more toward the back direction ($\theta = 180^\circ$). Fig.5.4. Shows the phase function of dust and water soluble aerosol at different scattering angle. The phase function for water soluble aerosol indicates stronger forward scattering as compared to that of dust aerosols. Generally, at visible wavelength water soluble aerosol are nonabsorbing. The more detail discussion

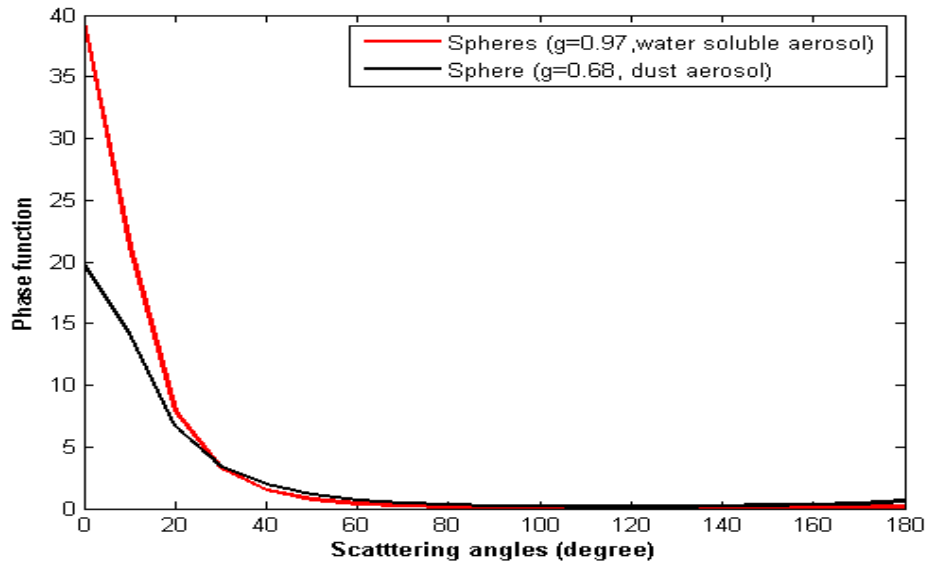


Figure 5.4: Comparisons of phase function for dust and water soluble aerosol

on different aerosol types on its phase function is given on reference [2].

Chapter 6

Conclusion

In this thesis, we modeled the aerosol particles as homogeneous spheres using the radiative transfer code based on Mie theory.

We simulate the aerosol optical thickness, which is one of the aerosol optical properties that are important in the atmospheric aerosol radiative properties. Aerosol optical thickness is an indirect measure of the size and number of particles in a given column of air.

To this end, we used an inversion technique, that we have developed to retrieve the particle size distributions from measured extinction data (aerosol optical depth) at six different wavelengths in the range between $0.34 \mu m \leq \lambda \leq 1.02 \mu m$. These were the only available for the data and time choosen for this study.

During a given inversion we assumed the aerosol particles are spherical, and the complex index of refraction of the particles are independent of wavelength and particle radius. However, we have varied index of refraction for a given aerosol type and repeated the experiment to see the sensitivity of columnar size distribution to the

assumed refractive index. We have also considered two aerosol type to determine if the degree of sensitivity between aerosols are different.

Results of our study reveal that particles with size radius comparable to shortest incident wavelengths contribute for higher fraction of attenuation of the electromagnetic radiation. On top of that, this result also shows that a change in refractive indices results in aerosol extinction spectra, that leads to changes in the retrieved aerosol size distribution. This observation is related to the fact that the refractive indices are dependent on chemical composition of the particles.

Our future investigation will be integrating the Ground-based aerosol optical thickness measurement with satellite based aerosol optical thickness measurements for a well advanced understanding of the aerosol size distributions in the Earth's atmosphere. Because of non-sphericity of most aerosols, we also plan to develop a retrieval algorithms which uses T-matrix theory for forward radiative transfer modeling.

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Declaration

I hereby declare that this thesis is my original work and has not been presented for a degree in any other university. All sources of material used for the thesis have been duly acknowledged.

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