The Scattering of Light by Non-spherical Particles

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Abstract

The following is a report of my activities in the second year of my D.Phil in atmospheric physics. The first chapter is an introduction to the subject. The second is a summary of most of my work this year which concerns the optical modelling of mineral dust aerosol. The third chapter covers a brief spell of work on the SPARCLE instrument which was left over from the first year of my D.Phil. Finally, a timetable and plan for future work is provided.
Chapter 1

Introduction

This chapter is a summary of the background physics used. Aerosol sources and their influence on the atmosphere are explored. Then, common terms in aerosol and light scattering physics are summarised.

1.1 What are aerosols?

An aerosol is a suspension of solid or liquid particles in a gas, generally with dimensions on length scales of nanometres to tens of microns. Initially, we can designate two categories: aerosols that arise from natural sources such as sand (mineral dusts) or emissions from volcanoes, and those that are from anthropogenic sources. This second category would mainly consist of the many products of fuel combustion and biomass burning (although these can also occur naturally).

Primary aerosol is composed of particles emitted into the atmosphere. Secondary aerosol would be those particles formed in the atmosphere by gas-to-particle conversion processes [Seinfeld and Pandis, 1998]. Concentrations can reach as high as several thousand per cubic centimetre for small particles ($< 1 \mu m$), whereas bigger particles have number densities less than 1 part. cm$^{-3}$.

Removing the particles from the atmosphere are two principal mechanisms: dry deposition (where the particles return to the Earth’s surface) and wet deposition (the particles are incorporated into cloud droplets which can precipitate out).

Distribution

Differences between aerosol in the troposphere and stratosphere are very apparent. Tropospheric content can vary in size, shape (from spherical, to extremely non-spherical) and refractive index. By contrast, stratospheric aerosol is, for the most part, aqueous sulphuric acid of varying concentrations. Volcanic eruptions can be a large source of SO$_2$ which enters directly into the stratosphere, and, after these events, we see the stratospheric levels of sulphate gradually decrease back to a background level with an e-folding time of months [Mergenthaler et al., 1995].
Distribution of the particles is generally characterised by a log-normal distribution with three principal modes. The smallest mode is nucleation ($5 \times 10^{-3}$ to $0.1 \mu m$), where particles are formed from the nucleation of atmospheric species in gas to particle conversion either on existing particles, or through condensation [Colbeck, 1998].

The second, and most populated mode, is the accumulation mode with particle diameters of 0.1 to $2.5 \mu m$. Here, we find particles which arise from the condensation of low-volatility vapours, and from coagulation of nucleation mode particles. Particles in this range are subject to the least number of removal mechanisms, so numbers accumulate here. These first two modes are collectively named the fine mode.

Finally, the coarse mode ($> 2.5 \mu m$) consists of large particles, formed by mechanical processes.
such as weathering and wind erosion. Due to their size, they have high sedimentation velocities (which increase roughly quadratically with size) and settle out of the atmosphere reasonably quickly (by dry deposition). The sources and sinks for the fine and coarse modes are different, and so we find very little exchanging of aerosols between the two.

Fig. 1.1 shows the three principal modes and their methods of growth and decay. Notice the one-way increase in the accumulation mode from the nucleation mode and the lack of exchange from fine to coarse particles.

1.2 Sources

Unlike most atmospheric gases, many aerosols have mixing times which are shorter than their atmospheric lifetimes. This leads to spatially inhomogeneous distributions of individual species. Consider the huge smog problems of Los Angeles where we find high concentrations of particulates, tropospheric ozone and NO$_x$ leading to low quality air and low visibility. Very steady meteorological patterns caused by the semi-permanent sub-tropical anticyclone over the Pacific ocean leads to descending air, high temperatures and very low wind speeds and inversion heights along the Californian coast. These conditions “trap” particulate in the cooler marine layer, where they are prevented from vertical exchange with the upper-air by the temperature inversion [Seinfeld and Pandis, 1998].

We can also see the non-homogeneous distribution of aerosols in Fig. 1.2 which shows total aerosol optical depth for two periods in 2001. It is immediately obvious that certain areas of the globe have higher concentrations of aerosol and often these are the industrial, continental regions. Increased optical depth also marks a clear plume extending into the Atlantic ocean from Saharan Africa and biomass emissions over the Amazon rainforests.

The following sections give a brief outline of aerosol sources. Particular attention is paid to those which are aspherical. It would be expected that the optical properties of these particles would be inferred badly in retrieval schemes which used the assumption of sphericity. Carbon chains and mineral dust cause the most problems because they are highly non-spherical. Although the categories “natural” and “anthropogenic” are not strictly accurate names, most species are created predominantly by one mechanism. The particulate that is mainly in the atmosphere due to human activity will be termed anthropogenic.

1.2.1 Natural aerosols

The sources of natural aerosols are controlled by local environmental conditions. Wind, temperature and humidity all play a part in affecting the rate of emission. Many of the particles are “kicked up” into the atmosphere so wind speed is the main factor determining population, and we see large temporal variation in natural aerosol. Of the natural aerosols, the significant non-spherical component is mineral dust.

$^1$For example, burning can also be because of natural wild fires.
Figure 1.2: Total aerosol optical depth determined from satellite measurements, showing seasonal and geographical changes in aerosol concentration. In white areas, no values are available due to the high reflectivity of mineral dust (in the Sahara) and snow (in the Arctic and Antarctic regions). Dots show locations of ground-based measurements, complementing the satellite data. This plot is Fig. TS.4. from the IPCC report [Solomon et al., 2007].
Salt

Marine aerosol is mainly composed of sea-salt. At the surface of the ocean, air bubbles burst, spraying droplets which either fall back to the surface of the water (if they are too large) or are carried higher, forming aerosol. Depending on the wind speed, the aerosol will or will not be able to reach the boundary layer (the faster the wind, the more likely) and we find that sea-salt concentration varies roughly linearly with wind speed [Colbeck, 1998]. Sea-salt aerosol is also responsible for sulphate formation (as an active site). The main salts are NaCl, KCl, CaSO$_4$ and Na$_2$SO$_4$ and atmospheric emission is estimated at 1300 Tg yr$^{-1}$ [Colbeck, 1998, Table 7.2]. This is a large proportion of the natural tropospheric aerosol but we find concentrations rapidly decrease as we travel inland from the oceans. Salt aerosol can be considered spherical as it will generally be suspended in water droplets.

Mineral dust

Mineral dust from desert and semi-arid regions is a major source of tropospheric aerosol. About 50% of this is subject to long-range transport and will turn up all over the world. Fig. 1.3 shows a dust plume extending out over the Atlantic ocean from the West coast of Africa; this is an example of the huge influence that mineral constituents can have on the optical properties of the atmosphere. Saharan mineral dust is thought to play a vital role in the support of vegetation in the Amazon basin and provides nutrient iron to the biogeochemical cycle of the ocean systems [Solomon et al., 2007].
Dust is brought into the atmosphere by saltation, a jumping motion whereby already detached, larger particles collide with obstructions on the surface bed, projecting smaller particles into the air. Interestingly, wind on its own is not enough to remove particles from the surface bed. Experiments carried out by Bagnold in the 1940s found that when a steady stream of air was blown over loosely scattered, fine cement, no particle motion was recorded, even at very high wind speeds [Bagnold, 1941]. Larger particles, already in motion, were required to lift the particulate. A good description of saltation is provided in Twomey [1977].

Since all of the fine mineral dust (which is light enough to be lifted into the troposphere) has been removed from the surface, most of the Saharan region does not provide huge sources of dust and, generally, the mineral dust is very coarse. Specific regions such as the Bodélé Depression in Chad, where annual deposition occurs, along with other preferable circumstances (including strong surface winds and favourable topography), provide the world with its mineral dust, this in fact being the world’s largest dust source [Washington et al., 2006]. Apart from the Sahara, the other major world source of mineral dust is the Gobi desert, with the US South West and the Saudi Arabian peninsula providing more local sources without the global reach.

The size of mineral dust in the atmosphere is around 1 $\mu$m and varies hugely in shape. Fig. 1.4 shows scanning electron microscopy of sample Saharan mineral dust\(^2\), demonstrating the sharp corners and varying shape which make these particles most certainly not spheres.

Although the mineral dust aerosol is mainly from desert regions, other types of soil can enter the atmosphere in a similar way to these sands, but come from different regions, having different consistency. We also need to consider the moisture of the surface, and the vegetation cover, both of which can prevent removal of particulate.

Principal constituents of mineral dust are oxides and carbonates of silicon, aluminium, calcium and iron. Table 1.1 gives the abundances of major soil constituent elements.

\(^2\)It is highly likely that this mineral dust sample was collected at ground level since the sizes are not representative of dust in the troposphere, being too large.
Volcanic emissions

Solid and gaseous material is injected into the atmosphere sporadically with the eruption of volcanoes. The larger solids are heavy and mostly deposited in the local area due to gravity, but huge amounts of ash, SO$_2$ and HCl are left in the atmosphere. Ash itself principally consists of SiO$_2$, Al$_2$O$_3$ and Fe$_2$O$_3$ and is in the coarse aerosol mode, mostly impacting on the regional scale. Globally, the main result of volcanoes is the creation of sulphate aerosol in the stratosphere. The explosive nature of eruptions means that sulphate can be injected buoyantly into the stratosphere where there are none of the precipitation effects that make sulphate lifetimes in the troposphere so much shorter. Thus residence times for stratospheric aerosols is estimated at 6–9 months [Colbeck, 1998].

The eruption of Mount Pinatubo on Luzon Island, Philippines in 1991 was the largest in recent times. Satellite measurements of column SO$_2$ estimated 20,000 kt was released and encircled the Earth within 22 days [Bluth et al., 1992]. As an example of the impact of these events, consider the year of the eruption. Records show that the warmest years in the 20th century have all taken place after the late 1960s. In 1991 and 1992 there was a sudden drop in global mean air temperature, due to the spread of aerosols in the stratosphere from the Pinatubo eruption [Bhutiyani et al., 2007], the only significant blip in an otherwise upward trend of global temperature. It took more than two years for aerosol quantities in the stratosphere to decay back to “normal” levels.

Since the sulphate aerosol is dissolved H$_2$SO$_4$, it can be assumed to be spherical. This is not the case for the ash, but we generally find that in periods not directly after an eruption there is not much of this in the air.

1.2.2 Anthropogenic aerosols

Anthropogenic aerosols come from particulate emissions from industry, transport and natural sources such as farming. In urban areas, the spread of sizes can be very variable, with sample sites close to a source (for example, a road) having extremely high concentrations of fine particulate. As we move away from the source, we see a rapid decrease in the concentrations of these smaller particles, as shown in Fig. 1.5.

The combustion of fuels also leads to secondary material from the chemical reactions in burning. These are gas-to-particle reactions in the atmosphere and the products are sulphates, nitrates and ammonium. Industry and agriculture are the main sources with mining, stone crushing in quarries and grain elevators all contributing heavily to coarse mode particles. However, the main area of interest from a non-spherical scattering perspective is black carbon, and other, secondary organics

<table>
<thead>
<tr>
<th>Element</th>
<th>Si</th>
<th>Al</th>
<th>Fe</th>
<th>Ca</th>
<th>Mg</th>
<th>Na</th>
<th>K</th>
<th>Ti</th>
<th>Mn</th>
<th>Cr</th>
<th>V</th>
<th>Co</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>330,000</td>
<td>71,300</td>
<td>38,000</td>
<td>13,700</td>
<td>6,300</td>
<td>6,300</td>
<td>13,600</td>
<td>4,600</td>
<td>850</td>
<td>200</td>
<td>100</td>
<td>8</td>
</tr>
</tbody>
</table>

Table 1.1: Average abundances of major mineral elements in soil and crustal rock, from Seinfeld and Pandis [1998]. Elemental abundances are in ppm by mass.
which are highly non-spherical.

**Carbon**

The main carbon molecules referred to are black carbon (BC), but organic molecules and a plethora of smaller carbon molecules created by combustion should also be mentioned.

The light-scattering properties of BC aggregates are tricky characteristics to investigate. Unfortunately, BC is one of the most prevalent anthropogenic aerosols. Table 7.1 from Chapter 7 of Colbeck [1998] shows that of all compounds and elements in the urban aerosol class, 29% by mass are organics (30.4 µg m\(^{-3}\) of total 103.6 µg m\(^{-3}\)). In fact, during the burning season, one third of the South African continent can be covered in smoke aerosol from biomass burning [Remer et al., 1998].

It should be mentioned that not all of the carbon in the atmosphere is from anthropogenic sources. Natural emissions from the biosphere also contribute to this group.

Biomass burning is very significant source of emissions. Again, some of these are from natural wild fires, but the largest contributor is the clearing of forest and savannah for agricultural purposes. The particles are incomplete combustion products, soot, sulphate, nitrate and hydrocarbons. Gases CO\(_2\), CO, CH\(_4\) and volatile organic compounds are also emitted. The type of fuel burnt makes a difference. Spark-ignition engines have high organic carbon content, while diesel engines emit more elemental carbon [Colbeck, 1998].

BC chains undergo a rapid evolution in the atmosphere due to coagulation, outgassing, condensation and gas-particle conversion. Starting as long, open chains, soot will gradually collapse into
densely packed clusters over a time period of hours [Abel et al., 2003]. Reid and Hobbs [1998] found that when measuring biomass aerosol very close to source, the single scatter albedo, $\bar{\omega}_0$, could be very low compared to the evolved chains which had spent more time in the atmosphere. Combustion with high BC content (30%) could have $\bar{\omega}_0$ at the limit set by Mie theory for highly absorbing particles with similar size scales. Knowledge of refractive index and particularly shape are not well known. Other properties such as mixing type of the suspension can result in differing absorbing of light so a better picture of these particles is very important. We can also find that the particles become coated by a non-absorbing shell such as organics or sulphates (forming an internal mixture) if the BC particles are formed at a high temperature [Martins et al., 1998].

A useful property of soot is that it is very persistent in the atmosphere, due to being inert and relatively hydrophobic. This makes it a good tracer for anthropogenic aerosol emission.

**Secondary anthropogenic emissions**

The main source of secondary emissions is oxidation of $\text{SO}_2$ and the various nitrous oxides ($\text{NO}_x$). About 50% of these species will undergo oxidation before being deposited. Atmospheric $\text{OH}^-$ radicals oxidise to $\text{H}_2\text{SO}_4$ and $\text{HNO}_3$. Sulphuric acid has low vapour pressure, and all of this thus becomes aerosol mass. Nitric acid is split between gas and aerosol phases. The $\text{SO}_2$ comes from fossil fuel emissions and is the major component of upper tropospheric and stratospheric aerosol. The secondary aerosol are in the fine mode and spherical.

### 1.3 The influence of aerosols on the atmosphere

Aerosol influence on the atmosphere is, at the time of writing, the largest unknown in estimates of radiative forcing. Fig. 1.6 shows the IPCC’s\(^3\) best estimate for global mean radiative forcings in 2005, with their 90% confidence intervals marked. We can see immediately that by far the widest confidence intervals are for the aerosol effects and that there is a low level of scientific understanding. This gives an important motivation for research into aerosol light scattering properties.

Aerosols reflect light back into space and act as cloud condensation nuclei (CCN), increasing the albedo. They also act as surface for other chemical reactions in the atmosphere to take place, for example the destruction of ozone in the troposphere and the nighttime formation of nitric acid. Their temporal variability, different types, and the number of different effects and reactions they influence makes them fiendishly difficult to model.

### 1.3.1 Radiative influence

Radiative forcing (RF) is a measure of how changes to the atmosphere affect the climate system. It can be defined as the net change in irradiance at the tropopause. A positive value implies that there

\(^3\)Intergovernmental Panel on Climate Change. See Solomon et al. [2007]
Figure 1.6: The estimates for global mean radiative forcings, presented by Working Group One of the IPCC. LOSU is the level of scientific understanding. Notice that the two largest error bars are those on the estimates of the aerosol direct and indirect effects. This is Fig. TS.5 in Solomon et al. [2007].

is more incoming than outgoing radiation\(^4\). In Fig. 1.6, we see that the aerosol RF has been split into two categories, the direct effect and the cloud albedo effect.

The direct effect is the scattering and absorption of light, altering the Earth’s radiative balance. If the aerosols are purely scattering, the effect is always a negative RF since some of the light destined to pass through an atmospheric layer is now reflected, and will pass back into space. For absorbing aerosols, the effect is more variable. Haywood and Shine [1995], using a simplified formula, show that radiative forcing will be negative only if the single scatter albedo, \(\bar{\omega}_0\) satisfies:

\[
\bar{\omega}_0 > \frac{2R_s}{\beta(1 - R_s)^2 + 2R_s},
\]  

where \(R_s\) is the surface reflectance and \(\beta\) is the backscatter fraction, the ratio of backscattered to

\(^4\)The IPCC defines RF slightly differently as the “rate of energy change per unit area of the globe as measured at the top of the atmosphere” since 1750 (the beginning of the industrial era).
forward scattered light, obtained from the phase function [Wiscombe and Grams, 1976]. Hence we find that over dark surfaces (forests or oceans), the effect is still negative while over bright surfaces (desert or snow or if the aerosol is above a cloud), there can be positive RF.

Short wavelength light rays are affected by all aerosol, leading to less irradiance at ground level. Long wavelength forcing is only appreciable if the scattering particles are large enough, and so has less of an effect.

The indirect aerosol effect refers to interaction with clouds and will be discussed in §1.3.3.

### 1.3.2 Chemical influence

Heterogeneous reactions involve the reaction of a gas molecule with a solid or liquid surface. This makes aerosols ideal atmospheric reactants in these cases.

In the troposphere for example, dry sea-salt acts to liberate hydrogen chloride when it comes into contact with gaseous nitric acid. Ozone destruction can also take place on the surface of carbon based aerosol, leaving us with CO and CO$_2$. Hazes in the troposphere offer wet surfaces for heterogeneous reactions, for example the nighttime formation of nitric acid.

In the stratosphere, polar stratospheric clouds, consisting of frozen or liquid sulphuric and nitric aerosol, are responsible for heterogeneous chlorine activation reactions which have a heavy role in polar ozone depletion [Colbeck, 1998].

### 1.3.3 Clouds

The indirect aerosol effect consists of several interactions which change the microphysical properties of clouds. Aerosols are vital in cloud formation, since they act as cloud condensation nuclei (CCN), and since clouds are a big influence on the Earth’s albedo due to their high reflectivity, changes in atmospheric aerosol concentration inevitably have an effect on RF (although, as Fig. 1.6 has demonstrated, an understanding of the subtleties is in no way complete).

The first indirect effect, or Twomey effect [Twomey, 1977], is an obviously direct consequence of increased particulate in the atmosphere: more CCN mean that there are more sites where droplets can form in developing clouds. This leads to greater numbers of droplets, but of a smaller size than in an unperturbed cloud (leading to an increase in the surface area of water droplets in the cloud). The end result for the radiative properties of the cloud is an increase in light reflected back into space and thus a negative radiative forcing.

Second indirect effects include the suppression of drizzle with greater numbers of CCN and other factors [Albrecht, 1989, Yia et al., 2008] which increase the liquid water content of the cloud. The cloud height can also increase, which leads to emission of radiation as the clouds rise and cool [Pincus and Baker, 1994]. The lifetime of the cloud is also increased since the reduced drizzle stops the cloud from breaking up [Albrecht, 1989].

A good overview of cloud effects can be found in §2.4.1 of the IPCC report [Solomon et al., 2007].
1.4 Aerosol characteristics

For a sample of spherical, atmospheric aerosol, only a small number of parameters are required to fully describe its optical properties. At the current time, assumptions are often made in retrieval algorithms to reduce complexity. In the ORAC\(^5\) scheme, for example, we firstly assume that the particles are homogeneous, and have a known refractive index (RI). Secondly, we build up our distribution from a number of lognormal aerosol modes. Assuming the number distribution of sizes, \(n(r)\), is lognormal, we require only the median radius, \(r_g\), and \(\sigma\) (the standard deviation of \(\ln r\)) to fully compute the ensemble’s light scattering properties. The lognormal distribution is defined in this case as:

\[
n(r) = \frac{1}{\sigma r \sqrt{2\pi}} \exp \left[ -\frac{1}{2} \left( \frac{\ln r - \ln r_g}{\sigma} \right)^2 \right]. \tag{1.2}
\]

The refractive index is then used, along with Mie theory, to calculate the light interaction properties of the aerosol. In this report, refractive index values and distribution properties of the various aerosol types are obtained using data, for the most part, from the Optical Properties of Aerosols and Clouds software package (OPAC) [Hess et al., 1998], but also from a study of Dubovik et al. [2002].

As soon as we remove our assumption of spherical, homogeneous particles, additional aerosol properties begin to appear. Most analysis of particle shape is only partially quantitative in that a sample of particulate is generally inspected under an electron microscope, and the shape properties of these few are manually measured. As such, we are only looking at a two dimensional image of the aerosol, and various parameters are used to characterise shape.

**Aspect ratio (AR)** The first is the AR, defined as the ratio of length along maximum projection of the shape and the width (perpendicular to that projection). This is very similar to the parameter \(\epsilon\) given in definition of spheroids and gives the ratio of \(a/b\), the major and minor axes, about one of which, the ellipse will be rotated to form our spheroid. For \(\epsilon = 1\) we have a sphere; \(\epsilon < 1\) implies a prolate spheroid (e.g. rugby ball); \(\epsilon > 1\) implies an oblate spheroid (e.g. a disk).

**Circularity (CIR)** Second, the CIR gives a measure of how a particle’s projected perimeter compares to its projected area. It is usually defined as \(\text{CIR} = \frac{\text{perimeter}^2}{(4\pi A)}\) (e.g. Kalashnikova and Sokolik [2004]) or the reciprocal of this expression. In some literature, it is called the shape factor, \(Sh\). As with AR, CIR should be unity for a perfect sphere.

**Fractal dimension** When looking at aggregate particles (such as fresh products of combustion), it is often true that their shapes are scale invariant [Sorensen, 2001]. In this case, a fractal dimension is a useful quantity. Essentially, a very similar quantity to circularity, it tells us how an agglomerate particle’s length relates to its mass. E.g., in Wentzel et al. [2003], they define

\(^5\)Oxford-RAL retrieval of Aerosol and Cloud
it as $D_f$ in the equation $N_p \propto \left( \frac{R_g}{r_p} \right)^{D_f}$, where $R_g$ is some radius of a particle describing overall size, and $r_p$ is the radius of an individual spherule within the whole.

There are also measurement parameters which take account of non-sphericity in three dimensions, such as the dynamic shape factor given by Kaaden et al. [2009]. Since these generally don’t provide information about specific shape (so much as about the correction required to the spherical case), they are not particularly useful in this case.

1.5 Optical Theory

1.5.1 Methods of calculating light scattering

Mie theory

Mie theory was developed by Gustav Mie [Mie, 1908] in order to understand the colours that resulted from light scattering from gold particles suspended in water. Although it is exact, only with the emergence of computing has it become practical to calculate the scattering values. An excellent introduction to the theory is provided in Bohren and Huffman [1983], whose notation and outline I have used. Code to calculate Mie scattering properties and their derivatives with respect to size and refractive index in the IDL programming language can be found on the AOPP, Oxford website\(^6\).

An incident, electromagnetic, $x$-polarised plane wave is expressed in vector spherical wave functions (VSWF), $M_{mn}(kr)$ and $N_{mn}(kr, \theta, \phi)$, which are solutions to $\nabla^2 E + k^2 E = 0$, in spherical coordinates. The incident electric field is shown to be

$$E_i = E_0 \sum_{n=1}^{\infty} \frac{i^n 2n + 1}{n(n+1)} \left( M_{1n}^{(1)} - i N_{1n}^{(1)} \right),$$

the subscripts $e$ and $o$ meaning only the even, and odd solutions respectively of $M_{mn}(kr)$ and $N_{mn}(kr, \theta, \phi)$ are wanted. The magnetic field can be simply obtained from $\nabla \times H = -i\omega \epsilon E$ where $\epsilon$ is the electric permittivity and $\omega$ the frequency. Applying boundary conditions at the sphere radius, $r_0$,

$$(E_i + E_s - E_I) \times \hat{e}_r = (H_i + H_s - H_I) \times \hat{e}_r = 0 \quad \text{for } r = r_0,$$

and writing the scattered field, $E_s$ and internal field, $E_I$, as a similar sum of VSWFs to our incident field, we find our scattered field solution, which turns out to be a sum of Riccati-Bessel functions and associated Legendre functions.

Coated spheres

For coated spheres, we only need to extend the theory of the previous section. Boundary conditions are imposed at the boundary between 1st and 2nd media as well as between the 2nd medium and

\(^6\)http://www-atm.physics.ox.ac.uk/code/mie/ Details of the code are given in Grainger et al. [2004].
the containing medium. After these are satisfied, we have a very similar solution to the Mie case. Details of this can be found in Bohren and Huffman [1983, pp181–183].

**T-matrix method**

The method used in the following work for non-spherical light scattering is the T-matrix method. This writes the incident field as a sum of VSWFs,

\[
E_i(r) = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \left[ a_{mn} M_{mn}^{(1)}(kr) + b_{mn} N_{mn}^{(1)}(kr) \right],
\]

the scattered field similarly,

\[
E_s(r) = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \left[ p_{mn} M_{mn}^{(3)}(kr) + q_{mn} N_{mn}^{(3)}(kr) \right],
\]

and the field on the surface of the scattering object as

\[
\hat{n} \times E(mkr) = \hat{n} \times \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \left[ c_{mn} M_{mn}^{(1)}(mkr) + d_{mn} N_{mn}^{(1)}(mkr) \right],
\]

where \( m \) is the refractive index and bracketed superscripts on the VSWFs denote specific solutions [Mishchenko et al., 2000], one can use the orthogonality properties of VSWFs to obtain relationships between just \( a_{mn} \) and \( b_{mn} \) or just \( p_{mn} \) and \( q_{mn} \) and the surface fields on our scattering object. These relationships can be written:

\[
\begin{bmatrix} a \\ b \end{bmatrix} = Q_1 \begin{bmatrix} c \\ d \end{bmatrix}, \quad \begin{bmatrix} p \\ q \end{bmatrix} = Q_2 \begin{bmatrix} c \\ d \end{bmatrix},
\]

and so we can relate the unknown scattering coefficients \( (p, q) \), to the known incident field coefficients \( (a, b) \) by

\[
\begin{bmatrix} p \\ q \end{bmatrix} = Q_2 Q_1^{-1} \begin{bmatrix} a \\ b \end{bmatrix},
\]

where the T-matrix, \( T = Q_2 Q_1^{-1} \). The matrices \( Q_1 \) and \( Q_2 \) are to be found, from which the scattered field can be easily calculated.

**1.5.2 Additional modeling methods**

The discrete dipole approximation (DDA) can in theory calculate the light scattering characteristics of almost any shape, at the price of much greater computational complexity. It also suffers from the disadvantage (not present in T-matrix or layered sphere methods) that for each incoming and
CHAPTER 1. INTRODUCTION

Figure 1.7: TEM images of typical black carbon aggregates, soon after their creation. From Kirchstetter and Novakov [2007].

outgoing light direction incident on the particle, the entire calculation must be repeated. In contrast, techniques which solve the wave equation as an expansion of vector spherical wave functions (VSWF) such as Mie theory or the T-matrix method, do not need to recalculate when reorientating their particles. In a system such as the atmosphere, where a huge distribution of randomly orientated particles are involved, this problem make DDA cumbersome.

A second group of methods concentrate on aggregate particles. There are basically two ways to proceed. One can look at individual particles, with a specific build up of spheres which represent a shape such as that shown in Fig. 1.7. These scattering solutions can be calculated exactly, although they are time consuming [Xu et al., 2004]. Aggregates can also be modelled as a statistical ensemble of fractal particles with a fractal dimension, $D$ [Sorensen, 2001].

1.5.3 Relevant Optical Properties

Size parameter

When looking at how light will scatter from a particle, the most important question is how comparable is the wavelength of light to the particle’s size. If the wavelength is much greater than the size of the scattering object, the object won’t be able to deflect the light in any significant way. As a result, the size parameter, $x$, is a clearer measure of the size regime in which we are interested than an absolute measure of the particle.

$$x = \frac{2\pi}{\lambda} r_0 = k r_0,$$  \hspace{1cm} (1.10)
where $\lambda$ is the wavelength of light and $r_0$ is the characteristic length of a particle and $k$ is the circular wavenumber. For a sphere, $r_0$, is the radius of the particle but for non-spherical particles, the choice of $r_0$ is not so clear. Mishchenko and Travis [1998] have written T-matrix code which gives users the option of defining $r_0$ as the radius of a sphere of either equal volume, or equal surface area to that of the scattering object.

**The amplitude matrix**

The amplitude matrix relates the scattered amplitude, $E_s$, to the incident amplitude of the electric field, $E_i$, where the light is split into light polarised parallel and perpendicular to the plane of scattering.

$$E_s = \frac{e^{ikr}}{kr} S E_i,$$

$$\begin{pmatrix} E_{\parallel s} \\ E_{\perp s} \end{pmatrix} = \frac{e^{ikr}}{kr} \begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} \begin{pmatrix} E_{\parallel i} \\ E_{\perp i} \end{pmatrix}.$$  \hspace{1cm} (1.11)

Definitions vary slightly between texts. For example, Bohren and Huffman [1983] multiply the prefactor by $i$, or Mishchenko et al. [2000] by $k$ (as well as using $E_\theta = E_{\parallel}$ and $E_\phi = -E_{\perp}$). Note the somewhat odd choice of element numbering in (1.11) which is due to convention in solving for the spherical case. This is a far field solution where it can be assumed that the scattered field is transverse ($E_s \cdot \hat{e}_r \simeq 0$). The elements of $S$ will be complex.

**Stokes parameters**

Stokes parameters are closely linked to the electric field, but have been chosen to mirror easily measurable scattering quantities. From van de Hulst [1957], we say:

$$I = \begin{pmatrix} I \\ Q \\ U \\ V \end{pmatrix}$$

where

$$I = E_{\parallel} E_{\parallel}^* + E_{\perp} E_{\perp}^*,$$

$$Q = E_{\parallel} E_{\parallel}^* - E_{\perp} E_{\perp}^*,$$

$$U = E_{\parallel} E_{\perp}^* + E_{\perp} E_{\parallel}^*,$$

$$V = i(E_{\parallel} E_{\perp}^* - E_{\perp} E_{\parallel}^*).$$  \hspace{1cm} (1.12)

The intensity is $I$ and the other elements measure the various amounts of polarisation. For a completely polarised beam, we have $I^2 = Q^2 + U^2 + V^2$ and for partially polarised beams, $I^2 \geq Q^2 + U^2 + V^2$. The degree of linear polarisation is given by $Q$ and $U$ and circular polarisation by $V$, as shown in Table 1.2.
Direction of polarisation for unit intensity ($I = 1$) & Stokes parameter

<table>
<thead>
<tr>
<th></th>
<th>Q</th>
<th>U</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unpolarised</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$\hat{e}_\parallel$</td>
<td>0°</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>$\hat{e}<em>\parallel$ ($\hat{e}</em>\parallel + \hat{e}_\perp$)/$\sqrt{2}$</td>
<td>45°</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>$\hat{e}<em>\parallel$ ($\hat{e}</em>\parallel - \hat{e}_\perp$)/$\sqrt{2}$</td>
<td>-45°</td>
<td>0</td>
<td>-1</td>
</tr>
<tr>
<td>Right circular</td>
<td>⬇️</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Left circular</td>
<td>⬆️</td>
<td>0</td>
<td>-1</td>
</tr>
</tbody>
</table>

Table 1.2: Showing the meaning of the values of different Stokes Parameters for polarised light. Adapted from Bohren and Huffman [1983].

Scattered parameters can be calculated from the incident parameters using the phase matrix, $Z$:

$$
\begin{pmatrix}
I_s \\
Q_s \\
U_s \\
V_s
\end{pmatrix} = \frac{1}{k^2 r^2}
\begin{pmatrix}
Z_{11} & Z_{12} & Z_{13} & Z_{14} \\
Z_{21} & Z_{22} & Z_{23} & Z_{24} \\
Z_{31} & Z_{32} & Z_{33} & Z_{34} \\
Z_{41} & Z_{42} & Z_{43} & Z_{44}
\end{pmatrix}
\begin{pmatrix}
I_i \\
Q_i \\
U_i \\
V_i
\end{pmatrix}.
$$

(1.13)

The values for elements $Z_{ij}$ (as a function of the amplitude matrix) are given as eqn (3.16) in Bohren and Huffman [1983] and eqns (13–29) in Chapter 1 of Mishchenko et al. [2000]. As with the definition of the amplitude matrix, slight differences occur. Mishchenko et al. [2000] multiply (1.13) by $k^2$ and have differing signs for some off diagonal elements, due to the use of unit vector $\hat{e}_\phi$ instead of $\hat{e}_\perp$.

**The phase function**

The phase function is a normalised measure of how the intensity of scattered light varies with scattering angle. For a single, spherical particle, irradiated with unpolarised incident light, this is defined as [van de Hulst, 1957, Bohren and Huffman, 1983]:

$$
p(\theta) = \frac{2}{C_{\text{sc}} k^2 r^2} \left( |S_1(\theta)|^2 + |S_2(\theta)|^2 \right),
$$

(1.14)

where $C_{\text{sc}}$ is the scattering cross-section and $S_1$ and $S_2$ are the amplitude functions for perpendicular and parallel components of the electric field. If we know the incident irradiance, $I_0$, the scattered intensity can be calculated as

$$
I = I_0 C_{\text{sc}} P(\theta).
$$

(1.15)
Asymmetry parameter

The asymmetry parameter is calculated from the phase function and tells us whether more light scatters in the forward or backwards direction.

\[ g = \langle \cos \theta \rangle = \frac{1}{4\pi} \int p(\theta) \cos \theta \, d\Omega. \]  

(1.16)

Positive values indicate more forwards scattering. If \( g = 0 \) then light scatters equally in all directions (or strictly, symmetrically about a scattering angle of \( \theta = 90 \)).

Cross-sections and efficiencies

Cross-sections tell us how much of an incident beam’s intensity is taken away\(^7\) in terms of the area of the beam. If the power taken away by a certain process is \( W_x \), then the cross-section for this process, \( C_x \), is:

\[ C_x = \frac{W_x}{\frac{1}{2} \sqrt{\frac{2}{\mu} |E_0|^2}}. \]

For example, scattering cross-section, \( C_{\text{sca}} \), tells us what area of the incident beam becomes scattered light. The extinction cross-section, \( C_{\text{ext}} \), tells us how much of the incident light is removed from the incident beam by scattering and absorption.

\[ C_{\text{sca}} = \frac{1}{I_i} \int_{4\pi} I_s(n_s) \, d\Omega, \]  

(1.17)

\[ C_{\text{ext}} = \frac{2\pi}{k} \Im \{Z_{11} + Z_{22}\} \]  

(1.18)

\[ C_{\text{abs}} = C_{\text{ext}} - C_{\text{sca}} \]  

(1.19)

where \( I_s = Z_{11} I_i + Z_{12} Q_i + Z_{13} U_i + Z_{14} V_i \) and \( Z_{ij} \) can be obtained from the Stokes parameters defined in (1.13).

Efficiencies are closely linked to cross-sections, but tell us what proportion of the beam incident on a particle is diverted to a certain process. As such, we define efficiencies, \( Q \), as:

\[ Q_{\text{ext}} = \frac{C_{\text{ext}}}{G}, \quad Q_{\text{sca}} = \frac{C_{\text{sca}}}{G}, \quad Q_{\text{abs}} = \frac{C_{\text{abs}}}{G}, \]  

(1.20)

where \( G \) is the cross sectional area of a particle, projected onto a plane perpendicular to the direction of incident light. In the case of a sphere of radius \( r_0 \), \( G = \pi r_0^2 \). Naively, we could guess that the extinction cross-section would be equal to \( G \), but since the influence of the particle outside the limits of its physical shape is non-zero, this is not the case.

---

\(^7\)The cross section is taken in a plane perpendicular to the direction of incident beam.
Single scatter albedo

The single scatter albedo tells us the probability that a photon incident on a volume element will survive.

\[ \bar{\omega}_0 = \frac{C_{\text{sca}}}{C_{\text{ext}}}. \]  

(1.21)

The range of values is \(0 \leq \bar{\omega}_0 \leq 1\).
Chapter 2

Improving the optical representation of sand

In this chapter, various parameters used in the definition of a desert dust aerosol class are modified to see how potential misrepresentations of aerosol would affect the optical properties observed. Namely, refractive index, shape and relative humidity of the surrounding atmosphere are changed.

2.1 Current state

The Oxford-RAL retrieval of Aerosol and Cloud algorithm (ORAC) forward model is a two-stage process [Thomas et al., 2005]. In the first stage, radiative properties (phase function, single scatter albedo, extinction) of the aerosol distribution are calculated for each satellite channel. In the second, a vertical profile of aerosol is generated, atmospheric gas absorption is calculated, and atmospheric transmission and bidirectional reflectance are obtained from the DISORT code [Stamnes et al., 1988].

The desert dust aerosol “class” is made up of four log-normal modes, taken from the Optical Properties of Aerosol and Clouds (OPAC) reference database [Hess et al., 1998]. The smallest of these modes is the “water soluble” mode, followed by the nucleation, accumulation and coarse mineral dust mode. The mineral modes are hydrophobic and the water soluble mode is assumed to always be present in an atmosphere with 50 % relative humidity. The three mineral dust modes all have identical refractive indices, the only difference between them being their size distributions.

Each mode has a defined median radius, $r_m$ and width $S$. The effective radius of the distribution, $r_{\text{eff}}$, is defined:

$$r_{\text{eff}} = \frac{\int_0^\infty r^3 n(r) \, dr}{\int_0^\infty r^2 n(r) \, dr},$$

where $n(r)$ is an arbitrary number distribution which is a function of individual particle radius, $r$. In order to alter $r_{\text{eff}}$ of the distribution, the size of particles, or their relative quantities can be changed. While $r_{\text{eff}}$ is greater than the effective radius of the smallest mode ($r_{\text{eff1}}$) and less than the effective
CHAPTER 2. IMPROVING THE OPTICAL REPRESENTATION OF SAND

Figure 2.1: Showing how the ORAC scheme generates aerosol distributions with the correct $r_{\text{eff}}$. Each colour represents a log-normal mode (or class). Blue is the “water soluble” class; green is the nucleation mode mineral dust; yellow the accumulation mode mineral dust; red the coarse mode mineral dust. The left hand figure shows the component mixing ratio for the four classes as a function of overall $r_{\text{eff}}$. On the right we see the median radius of each class as $r_{\text{eff}}$ varies. Overlaid crosses are microphysical measurements taken in the SAMUM campaign between 18th May and 7th June, 2006 in Casablanca, Ouarzazate and Zagora [Weinzierl et al., 2009, Tables 4 & 5].

radius of the largest mode ($r_{\text{eff4}}$), only the mixing ratio is altered\(^1\). When $r_{\text{eff}} < r_{\text{eff1}}$, it is assumed only the smallest class is present, and $r_{\text{eff}} = r_{\text{eff}}$. Similarly, for $r_{\text{eff}} > r_{\text{eff4}}$, only the largest class is present, and $r_{\text{eff4}} = r_{\text{eff}}$.\(^2\) This is shown in Fig. 2.1, along with measurements made in the recent SAMUM campaign in North-West Africa [Weinzierl et al., 2009]. In these measurements, a four mode size distribution of aerosol particles was retrieved from data provided by several particle sizers operating over a wide range of aerosol radii using the method of Fiebig et al. [2005]. This fit does not make assumptions about the chemical composition of the particles in each mode, so different coloured crosses denote only the size order for each measurement. A fairly good fit between the size distribution model and measurements gives confidence that this will not be a significant source of error in a forward model.

2.1.1 Comparing with field measurements

Having a size distribution of aerosols, we now model their optical properties. In the light scattering calculations, the main assumption made about the ORAC desert dust class is that the scattering particles are spherical and homogeneous. This allows us to use Mie’s solution to Maxwell’s equations [Mie, 1908] which are easily calculable and well known. This is certainly not the most sensible shape as any inspection of sand with electron microscopy will show you [e.g. Buseck et al., 2000,

\(^1\)I.e. $r_{\text{eff}}$ is made smaller by increasing the relative amounts of the smaller modes and vice versa.

\(^2\)For a lognormal distribution, $r_{\text{eff}} = r_m e^{-\frac{5}{2} \left(\ln S\right)^2}$
Okada et al., 2001, Reid et al., 2003, Kalashnikova and Sokolik, 2004]. Data have been inspected from several Saharan campaigns: SHADE [Tanré et al., 2003], DABEX [Haywood et al., 2008], DODO [McConnell et al., 2008] and, most recently, SAMUM [Heintzenberg, 2009]. Where values of extinction coefficient ($\beta_{\text{ext}}$), asymmetry parameter ($\langle \cos \theta \rangle$) and single scatter albedo ($\bar{\omega}_0$) could be derived, they have been inserted into Fig. 2.2 which shows these optical properties as they are currently represented in the forward model.

![Figure 2.2: Comparing available measurements from field campaigns presented in the literature to the current ORAC optical scattering properties table for $\lambda = 550$ nm. The references from which data was obtained are shown in the legend. Measurements shown with a cross (×) instead of a plus (+) do not include the coarse fraction of mineral dust.](image)

It would appear that in most cases $\beta_{\text{ext}}$ is underestimated by the forward model. Similarly, $\bar{\omega}_0$ is underestimated quite dramatically suggesting that the optical model is too absorbing. Happily, $\langle \cos \theta \rangle$ appears to be well represented over the range, although the range of values has a very small spread and this parameter is not actually used in retrievals (only as a single value representation of the phase function).

Certain caveats apply to the use of each set of field measurements for a fair intercomparison, and
these are listed below:

**Esselborn et al. [2009], Weinzierl et al. [2009]** The ORAC tables use $\beta^{\text{ext}} \cdot 10^{-3} \text{km}^{-1}$ for 1 particle per cm$^3$. In order to plot like with like, we need to know the number density (by which to divide the measured $\beta^{\text{ext}}$) and $r_{\text{eff}}$ (for the abscissa). These values were taken from Weinzierl et al. [2009, Tables 4 & 5] in both cases (since both papers were from the same campaign flights) while the measurements of $\beta^{\text{ext}}$ appeared to be for same flights at same (or similar) locations. Times of $\beta^{\text{ext}}$ measurements could vary by up to an hour from the size distribution measurements, and were obtained by sight from graphs of lidar measurements since data were not requested. Times coincided more closely for the $\beta^{\text{ext}}$ measurements made by Weinzierl et al. [2009]. Lidar measurements of extinction coefficient are made at 532 nm and calculated by comparing a “background” molecular vertical profile to the current signal. The above points could all contribute errors to the $\beta^{\text{ext}}$ provided in Fig. 2.2.

**Curtis et al. [2008]** These measurements of $r_{\text{eff}}$ and \langle \cos \theta \rangle are taken from measurements of individual types of dust e.g. calcite or gypsum as opposed to a specific blend of minerals expected to be in real desert dust.

**McConnell et al. [2008]** In this flight campaign, $\bar{\omega}_0$ is measured, and $r_{\text{eff}}$ is obtained from values of $r_m$, $S$ and mixing ratio for 4 mode lognormal distributions given in Table 5. It is noted in the table that these size distributions are “Accumulation Mode Only”. Also, \langle \cos \theta \rangle is calculated by fitting a best refractive index to the measured size distribution to match the measured $\bar{\omega}_0$, assuming spherical sand particles. For values of $\beta^{\text{ext}}$, the measurements provided are mass specific extinction, $k^{\text{ext}}$ (in m$^2$ g$^{-1}$), derived from Mie fitting of $\bar{\omega}_0$ as for the \langle \cos \theta \rangle values. These are converted to $\beta^{\text{ext}}$ by multiplying by the value of sand density used in this study ($\rho = 2.65$ g cm$^{-3}$).

**Osborne et al. [2008]** As McConnell et al. [2008] above, but with a five-mode distribution. Measurements of $k^{\text{ext}}$ were provided with and without the coarse mode. Both values are plotted with the lower value being that which includes the larger particles.

**Haywood et al. [2003]** As Osborne et al. [2008] above, but with two different five-mode distributions provided. The values of \langle \cos \theta \rangle and $\bar{\omega}_0$ are calculated for the two assumed distributions (but are from the same data). As with Osborne et al. [2008], two values for $k^{\text{ext}}$ are provided.

### 2.2 Refractive index

One could suspect that across the world (and indeed within particular regions), variation in the minerology of sand and, by extension, the minerology of sand aerosol would be significant. Certainly, we would then expect the refractive index (RI) of the mineral dust aerosol to vary. Whether or not this causes unacceptable errors in calculations of optical properties is the next question. Claquin et al.
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Figure 2.3: “Damping” the imaginary part of refractive index, $k$, to see its effects on optical properties compared to measured properties as previously shown in Fig. 2.2. ($\beta^{\text{ext}}$, which has not been shown, is identical for all four cases.)

Figure 2.4: Different refractive indices used by the ORAC retrieval and Claquin et al. [1998].

[1998] reported that the imaginary part of refractive index, $k$, has “natural variability . . . [which can] lead to variations of up to ±40% in aerosol forcing calculations”, which suggests that these problems cannot be ignored. Looking again, for example, at Fig. 2.2, one could say that comparing the field measurements of $\bar{\omega}_0$ to the ORAC scheme, there is too much absorption in our representation of the optical properties of mineral dust at $\lambda = 550$ nm. This is equivalent to saying that the imaginary part of the refractive index has been over estimated.

As a naive first attempt, the absorption could be “damped” by reducing $k$ by an arbitrary amount. This is shown in Fig. 2.3. Reducing $k$ to between one half and one tenth of its original value would appear to provide a better fit to $\bar{\omega}_0$ while leaving $\langle \cos \theta \rangle$ similar to before, and $\beta^{\text{ext}}$ unchanged.

This uncertainty in $k$ was also inspected by Claquin et al. [1998] who used two different models of absorption for their mineral dust aerosol: a “close to source” containing more quartz, and “remote oceanic” which was more clay dominant. Fig. 2.4 shows how these two models compare to the ORAC
values in the visible channels. It can be seen that using the “close to source” values for $k$ would reduce the absorption over most of the visible spectrum (although only by a small amount at $\lambda = 550$ nm). The optical model used by OPAC appears to have more in common with the “remote” sand type, which could explain why aircraft measurements taken over the Sahara appear to have less absorption than our model predicts.

In an attempt to better understand and characterise uncertainty in optical properties due to the uncertainty in mineral dust refractive index, a spread of world wide mineral dust RI was calculated.

### 2.2.1 Method

Obtaining the mean RI was a three step process. First an approximate soil composition was obtained on a regular grid over the whole earth, built of common minerals. Next, the RI values of the individual minerals were combined for each soil type, giving a picture of how RI varies geographically. Finally, the worldwide spread was reduced to a histogram of RI values. These were weighted using a simple emissions scheme so that only areas where dust was likely to be lofted into the atmosphere contributed to the average.

**FAO / UNESCO Digital Soil Map of the World**

In order to characterize the spread of worldwide sand RI values, it was decided that soil data would be the most useful starting data set. The FAO\textsuperscript{3}/ UNESCO\textsuperscript{4} Digital Soil Map of the World [UNESCO, 2003] is a regularly gridded $5 \times 5$ arc-minutes resolution map of the world, each cell containing a mix of soil types. These soil types consist in varying proportions of different minerals.

Claquin et al. [1999, Table 2] provide a list of FAO soil types found in arid areas with the normalised composition by weight of important (sand) minerals, in this case illite, kaolinite, smectite, calcite, quartz, felspath (or felspar), gypsum and hematite. The minerals are divided into those found in clay fraction ($< 2 \, \mu m$) and the larger silt fraction ($2 \rightarrow 50 \, \mu m$). There is an even larger, sand fraction ($50 \rightarrow 200 \, \mu m$), but this was considered too large to be lofted for extended periods. Unfortunately, the FAO soil map uses ‘textures’ of soil (coarse, medium or fine) in order to characterize the sizes, so in order to estimate the mineral composition, a method is required to convert soil textures into soil sizes. Data from Zobler [1986] can help by giving approximate compositions for these textures, but these definitions are from wet sieving techniques which dissolve soluble minerals and dis-aggregate the soil, leading to likely over-representation of the smaller clay particles. Data from dry sieving studies contain much smaller amounts of clay which have a more “natural” distribution. Comparisons of size fractions for the two techniques are shown in Table 2.1. It was decided that a size distribution based on dry sieving techniques would be more useful. Since the “coarse sand” class shown above contains no clay particles at all, it was decided to use the next largest, “Coarse-medium sand” class which contained a very small fraction of clay (0.4 %).

\textsuperscript{3}Food and Agriculture Organization of the United Nations

\textsuperscript{4}United Nations Educational, Scientific and Cultural Organization
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<table>
<thead>
<tr>
<th>FAO textural class</th>
<th>Soil ratio (%)</th>
<th>Wet sieving</th>
<th>Dry sieving</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Clay</td>
<td>Silt</td>
</tr>
<tr>
<td>1 Coarse</td>
<td>9</td>
<td>8</td>
<td>83</td>
</tr>
<tr>
<td>1,2 Coarse-Medium</td>
<td>20</td>
<td>20</td>
<td>60</td>
</tr>
<tr>
<td>2 Medium</td>
<td>30</td>
<td>33</td>
<td>37</td>
</tr>
<tr>
<td>3 Fine</td>
<td>67</td>
<td>17</td>
<td>17</td>
</tr>
</tbody>
</table>

Table 2.1: Showing how selected textural classes can be described in terms of proportions of clay, silt and sand. Wet sieving data is from Zobler [1986, Fig. 2]. Dry sieving data is from Laurent et al. [2008, Table 4]. Notice that fractions of clay particles are much greater when wet sieving techniques are used to analyse the soil.

For each 5′ × 5′ area of land, the FAO map gives up to 8 soil types which make up a proportion of the land area, each with their own soil texture. Of course, not all of these soils will be suitable for lofting into the air.

**Converting soil types into refractive indices**

As mentioned above, the mineralogical breakdown of several soils was provided in Claquin et al. [1999, Table 2] who named 24 soils from the FAO soil map which were appropriate arid soil types. These were I, Jc, Je, Qa, Qc, Qf, Qi, Rc, Re, So, Vc, Xh, Xk, Xl, Xt, Xy, Yk, Yl, Yy, Zg, Zo, Zt, sand dunes and salt flats. For example, the cambic arenosol class, Qc21-1a which is found in North Africa consists of:

<table>
<thead>
<tr>
<th>Soil type</th>
<th>Clay fraction</th>
<th>Silt fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ill</td>
<td>Kao</td>
</tr>
<tr>
<td>Qc</td>
<td>30  %</td>
<td>16</td>
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<tr>
<td>Ql</td>
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<td>10</td>
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<tr>
<td>Re</td>
<td>20  %</td>
<td>32</td>
</tr>
<tr>
<td>Gc</td>
<td>10  %</td>
<td>30</td>
</tr>
<tr>
<td>Lk</td>
<td>10  %</td>
<td>37</td>
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<tr>
<td>So</td>
<td>10  %</td>
<td>32</td>
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</tbody>
</table>

Weighting the RI of each mineral, (using the appropriate clay and silt fractions for each soil type) we should then be able to work out the bulk refractive index of soil type Qc21-1a. Non-mineral-dust soil types are not included in the averaging.

A literature review was undertaken to track down the RI values at AATSR wavelengths of the minerals mentioned above, out of which Claquin et al. [1999, Table 2] built their sand types. The

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5Details on the various FAO soil classifications can be found at [http://www.fao.org/waicent/Faoinfo/agricult/agl/agll/key2soil.stm](http://www.fao.org/waicent/Faoinfo/agricult/agl/agll/key2soil.stm)
<table>
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<td>Illite</td>
<td>0.56</td>
<td>Egan and Hilgeman [1979]</td>
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<td></td>
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</tr>
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<td></td>
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<td></td>
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<td>Roush et al. [1991]</td>
</tr>
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<td>12.05</td>
<td>Egan and Hilgeman [1979]</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Roush et al. [1991]</td>
</tr>
</tbody>
</table>

Table 2.2: Source of refractive index values used in this study at the wavelengths of AATSR channels. Calcite values in the visible are assumed non-absorbing ($k = 0$). Calcite and quartz are birefringent. Since we expect any sand particles we observe to be randomly orientated, we combine the o-ray and e-ray values of $n$ and $k$, with twice the weighting for the o-ray values.

source for this data is given in Table 2.2. Additionally, the resolution of soil data is reduced from a $5' \times 5'$ grid to a $1.4' \times 1.4'$ grid to speed up further computations, reduce statistical noise in RI values, and reduce areas of unknown RI.

**Weighting by likelihood of emission**

The aim in this final stage is not to obtain a highly complex, detailed view of worldwide mineral dust emission. Instead, a basic scheme is developed which depends on the clay fraction of the mineral dust in a grid box, and a measure of peak wind speed at the same position, and is very quick to calculate with already available data.

The vertical emission flux of fine particles, $F$, is linearly related to the saltation flux, $G$ (the flux of particles travelling parallel to the soil surface), by the sand blasting efficiency, $\alpha$, so that we have,

$$ F = \alpha G. \quad (2.2) $$

From Laurent et al. [2008], we obtain $\alpha$ using the empirical relation

$$ \alpha = 10^{(0.134 \times \text{age clay} - 6)}, \quad (2.3) $$

but the saltation flux is more complex and will be approximated. The full calculation is [Marticorena et al., 1997]:

$$ G = E_D \rho_a u^3 \int_{D_p}^{\infty} \left(1 + \frac{u^*_p(D_p)}{u^*} \right) \left(1 + \left[\frac{u^*_p(D_p)}{u^*} \right]^2 \right) \delta S(D_p) dD_p, \quad (2.4) $$
where $E$ is the erodible fraction of the surface, $\rho_a$ is the air density, $g$ the acceleration due to gravity, $u^*$ the wind friction velocity, $u^*_t(D_p)$ the threshold wind friction velocity which gives the minimum value of $u^*$ needed to lift a particle of diameter $D_p$ from the surface. The size distribution is given by $\delta S(D_p)$.

Since we care about events when the wind speed is large relative to the threshold, we approximate (2.4) to:

$$G = \frac{F}{\alpha} \simeq u^3 f(D_p),$$

$$\Rightarrow \quad F \propto u^3 \alpha. \quad (2.6)$$

Wind friction velocity can be defined as [Almeida et al., 2006]:

$$u^* = u(z) \kappa \ln \left( \frac{z}{z_0} \right), \quad (2.7)$$

where the vertical distance, $z = 10$ m, $\kappa = 0.4$ is the von Karman constant and $z_0$ is the surface roughness length, which affects emissions, but is not trivial to calculate from the data we have obtained. In the emitting areas of the Sahara, the range of $z_0$ is from $z_0 = 10^{-3}$ cm to 0.5 cm so can alter $u^*$ by a factor of about 3. Due to the difficulty in obtaining $z_0$, it was decided to assume that $\ln \left( \frac{z}{z_0} \right)$ was almost constant relative to $u(z)$, the horizontal wind speed at 10 m.

Peak horizontal wind speed was obtained from a year’s worth of ECMWF data. The horizontal wind speed mean and standard deviation were calculated, and peak wind speed was defined as two standard deviations above the mean wind speed for each location.

The final concern is vegetation. If it is present, it all but completely inhibits emission of mineral dust. Tegen and Fung [1995] used a threshold based on the normalised difference vegetation index, $\text{NDVI} = \frac{R_{\text{nIR}}}{R_{\text{nIR}} + R_{\text{red}}}$, where $R_{\text{nIR}}$ and $R_{\text{red}}$ are the reflectances in the near-infrared and red regions respectively. Their approach was to allow emission only for $\text{NDVI} < 0.07$. A value of NDVI above this would suggest a vegetation cover “dense enough to inhibit dust deflation”. The AATSR monthly mean of atmospherically corrected NDVI for March 2004 was used as our data set [Sayer, 2008]. It was found that setting the cutoff at $\text{NDVI} < 0.08$ gave a more convincing emissions pattern. The AATSR NDVI product uses different bands for its nIR and red reflectance (which are also much narrower) than the bands from AVHRR used by Tegen and Fung [1995].

Since the NDVI index only shows an absence of vegetation, we must be careful not to mistake other bright areas (such as snow caps) for arid soil locations. Limiting the arid regions to between latitudes of 45°S and 45°N was a simple first step in this.

Fig. 2.5 shows the approximate calculation of relative emissions using the method described above for Saharan Africa, and compares it to a full, far more detailed calculation carried out by Laurent et al. [2008]. Although one could not state that a perfect match has occurred, there are very encouraging features of the stripped down emission scheme. Major known sources, for example
along the West Saharan coast, in central Algeria ($27^\circ$ N, $2^\circ$ W), and the Bodélé depression in Chad ($17^\circ 55'$ N, $19^\circ 7'$ E) are clearly visible in both representations. Many areas with no emissions at all also match, e.g. ($29^\circ$N, $2^\circ$E) but the region to the East of the Red Sea is disappointing. The simple model predicts quite a few emissions which are completely absent from the full calculation. The most likely reason for this would be soil moisture, which strongly restricts emission, but isn’t accounted for in our scheme.

### 2.2.2 Results

Fig. 2.6 shows predicted emissions for the whole world. Noticeable emitting regions are the Sahara, central Asia, central Australia, Nevada and the Chilean coast. As well as weighting an area on its emitting strength, because the data are on a latitude-longitude grid, areas close to the equator have greater surface area which must be accounted for. Coastal areas also have to account for loss of ground from which to emit due to parts of the grid being sea. This has all been added to the final calculations shown in Fig. 2.7–2.8 and summarised in Table 2.3 which shows optical characteristics.
of a mineral dust aerosol class with $r_{\text{eff}} = 1.13 \, \mu m$.

Values of $n_{550}$ seem slightly larger than previous studies [Highwood, 2009] which is a cause for concern but the absorption centering around $k_{550} = 1 \times 10^{-3}$ agree with other studies and are less absorbing than OPAC values, both of which are reassuring characteristics. Looking only at the Saharan region, we find $n_{550} = 1.58$ which is a slight improvement. New optical calculations based on the histograms of RI shown in Fig. 2.7-2.8 show (as one would expect) that with lesser values of $k$, $\bar{\omega}_0$ is greater than previously. Changes in $\langle \cos \theta \rangle$ are less pronounced, however the phase function can change by significant amounts in the very narrow backscatter direction of $\theta > 160^\circ$. For the backscattered light, $P_{550}(180^\circ)$, relative difference can be as great as 15%. Very little change is seen in the extinction.

Fig. 2.10 shows the spread of optical properties which is averaged to create Fig. 2.9. The values of $k_{\text{ext}}$ which are not shown vary only by very small amounts. For larger particles, variation in $\langle \cos \theta \rangle$ can reach 5% at $r_{\text{eff}} = 10 \, \mu m$ and variation in $\bar{\omega}_0$ can be up to 1.5%.

Almost all of this variation is caused by the imaginary part of RI. Holding $n$ constant while varying $k$ yields very similar results to allowing the full range of both $n$ and $k$. Conversely, holding $k$ constant while allowing $n$ to vary reduces greatly the variation in optical properties seen.

The conclusion would be that while imaginary part of refractive index is very important for the correct characterization of optical properties, particularly $\bar{\omega}_0$, the small variations in the real part (within the range shown by the soil analysis) do not greatly affect most parts of the phase function, or the other optical properties. The variability found in $k$ is not as large as the difference between OPAC values of $k$ and these new calculations. Incorrect characterisation of the imaginary part of refractive index is a greater concern than the natural variability in $k$ seen across different soil types which may be lofted into the atmosphere.

---

6For the purposed of this report, the relative variations reported are defined to be the standard deviation of values as a fraction of their mean value.
<table>
<thead>
<tr>
<th>WL (µm)</th>
<th>n</th>
<th>k × 10^{-5}</th>
<th>ORAC</th>
<th>Mean</th>
<th>SD</th>
<th>ORAC</th>
<th>Mean</th>
<th>SD</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.55</td>
<td>1.53</td>
<td>1.59</td>
<td>0.018</td>
<td>550</td>
<td>118.00</td>
<td>51.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.66</td>
<td>1.53</td>
<td>1.58</td>
<td>0.018</td>
<td>440</td>
<td>8.45</td>
<td>2.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.863</td>
<td>1.53</td>
<td>1.57</td>
<td>0.018</td>
<td>400</td>
<td>11.90</td>
<td>3.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.59</td>
<td>1.53</td>
<td>1.56</td>
<td>0.018</td>
<td>595</td>
<td>9.16</td>
<td>3.9</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 2.3: RI variation and its affect on the full mineral dust aerosol class. The calculated RI values are compared to the OPAC values currently used by the ORAC retrieval. The class has four modes, but since the smallest mode is the OPAC water soluble mode, it has not been affected by these calculations. The other three modes have had their refractive indices altered to match the results shown in Fig. 2.7-2.8. The relative variances (RV) quoted are the SD divided by the mean.
Figure 2.6: The soil refractive index for arid soil types over the earths surface is shown, followed by the various parameters required to calculate the relative emitting strength of an area of the world. NDVI data are the monthly mean from March 2004 AATSR data. An area is only allowed to emit if its NDVI value is less than 0.08. Sand blasting efficiency, $\alpha$, is only calculated for the soil types which are mineral dust as defined by Claquin et al. [1999] and is obtained as a function of the clay fraction, using eqn (2.3). Peak horizontal wind speed is from ECMWF reanalysis data. The final result, normalized emitting strength, is a function of the previous plots.
Figure 2.7: Showing the spread of the real part of mineral dust aerosol refractive index for AATSR wavelengths.
Figure 2.8: Showing the spread of the imaginary part of mineral dust aerosol refractive index for AATSR wavelengths.
Figure 2.9: Comparing values of $\langle \cos \theta \rangle$ and $\bar{\omega}_0$ at 550 nm, calculated using the OPAC refractive indices, and using a weighted spread of RI from the histograms in Fig. 2.7-2.8. Two example phase functions are also shown. Crosses are the data previously shown in Fig. 2.2. New values of RI appear to better reproduce the SSA.

Figure 2.10: Showing the spread of optical properties from the mean value at 550 nm, using the weighted spread of RI from the histograms in Fig. 2.7-2.8. The solid line is the overall scattering as shown in Fig. 2.9 above. The colours show the density of points within a grid box. Dashed lines are the extreme values given by the range of $n$ and $k$. 
CHAPTER 2. IMPROVING THE OPTICAL REPRESENTATION OF SAND

2.3 Non-sphericity

A great number of studies have investigated the shape of mineral dust aerosol and a great many more, how best to model it. These studies are summarised, followed by a description of a non-spherical (NS) model for the mineral dust aerosol class. The new scattering model is used as the mineral dust class for a retrieval of a known dust event off the coast of northwest Africa and compared to the standard spherical retrieval.

2.3.1 Other studies

Certain characteristics are used to generalise the classification of aerosol shape. A literature review was undertaken and the results of this have been split into studies of measurement of mineral dust aerosol, and those which endeavour to model the non-sphericity in light-scattering calculations. Summaries of the various terms are provided in §1.4-1.5.

Measurement summary

Many campaigns have used images from scanning electron microscopy (SEM) and transmission electron microscopy (TEM), which provide 2D images. As such, it is almost always the projected features of a particle onto an image plane that are analysed. Studies where the shape of the particle was not analyzed have not been included. Scattering methods employed in these studies include Mie theory, the discrete dipole approximation (DDA), T-Matrix theory and ray tracing.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Location</th>
<th>Method</th>
<th>Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volten et al.</td>
<td>Dust from various worldwide locations.</td>
<td>Analysed using SEM and measurement of phase function.</td>
<td>-</td>
</tr>
<tr>
<td>[Buseck et al., 2000]</td>
<td>Asian dust</td>
<td>TEM analysis.</td>
<td>Mean AR $\sim 1.5$.</td>
</tr>
<tr>
<td>[Okada et al., 2001]</td>
<td>Three arid regions in China.</td>
<td>SEM analysis.</td>
<td>Mean AR $\sim 1.3 \rightarrow 1.6$. CIR decreased with increasing radius, suggesting aggregate particles at larger sizes.</td>
</tr>
</tbody>
</table>
Table 2.4 continued . . .

<table>
<thead>
<tr>
<th>Reference</th>
<th>Location</th>
<th>Method</th>
<th>Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Haywood et al., 2003]</td>
<td>Sahara (SHADE experiment)</td>
<td>Measurements of dust by Met Office C-130 aircraft. Including Small Ice Detector (SID) and Passive Cavity Aerosol Spectrometer Probe (PCASP).</td>
<td>Asphericity factor (af) measured by SID is greater than 1, suggesting non-spherical scattering patterns when in the dust aerosol layer.</td>
</tr>
<tr>
<td>[Reid et al., 2003]</td>
<td>Puerto Rico (PRIDE experiment).</td>
<td>SEM analysis of airborne measurements.</td>
<td>Median $AR = 1.9$ with standard deviation $0.9$ independent of size. Except some of the largest particles ($&gt;10\mu m$, $13%$ of samples) are slightly elongated ($AR = 2.2 \pm 1.2$).</td>
</tr>
<tr>
<td>[Kalashnikova and Sokolik, 2004]</td>
<td>Representative Saharan and Asian dust samples.</td>
<td>SEM images analysed and circularity, non-sphericity and AR calculated. These are compared to values for known shapes.</td>
<td>Various shape distributions found. From these, models are created (see Table 2.5).</td>
</tr>
<tr>
<td>[Kandler et al., 2007]</td>
<td>Saharan mineral dust measured at Izaña, Tenerife.</td>
<td>Automated analysis of 22,000 particles from SEM images.</td>
<td>Above $1\mu m$, the aerosol is dominated by mineral dust. Most particles have a coating of sulphates with average thickness $60\mu m$. Formula for distribution of $AR$, $h(AR)$ provided (modified log normal). Average $AR = 1.64$. $RI = 1.59 + 0.007i$.</td>
</tr>
</tbody>
</table>
Table 2.4 continued . . .

<table>
<thead>
<tr>
<th>Reference</th>
<th>Location</th>
<th>Method</th>
<th>Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Chou et al., 2008]</td>
<td>Niger, African Monsoon Multidisciplinary Analyses (AMMA)</td>
<td>FAAM flights, TEM and SEM. Aspect ratio (AR) calculated by comparing projected area to projected major axis from image for 790 particles.</td>
<td>AR is independent of size. Biomass burning particulate observed in fine fraction of mineral dust. Soot chains were not observed. “mineral dust particles could be described as ellipsoids” whose major axis never exceeds 2.2 times the minor axis.</td>
</tr>
</tbody>
</table>

Modelling summary

Table 2.5: Modelling of dust properties

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>Assumed properties</th>
<th>Conclusions</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Mishchenko et al., 1997]</td>
<td>T-matrix [Mishchenko and Travis, 1998].</td>
<td>“Equiprobable shape mixture of prolate and oblate spheroids” with AR ranging from 1.4 → 2.2 in steps of 0.2.</td>
<td>$P_{\text{NS}} \gg P_{\text{S}}$ for $90^\circ \leadsto 150^\circ$; $P_{\text{NS}} \ll P_{\text{S}}$ for $150^\circ \leadsto 180^\circ$, where $P_{\text{S}}$, $P_{\text{NS}}$ are spherical and non-spherical (NS) phase functions.</td>
</tr>
<tr>
<td>[Wang et al., 2003]</td>
<td>T-matrix [Mishchenko and Travis, 1998]</td>
<td>Composite phase fn $P(\theta) = \frac{1}{3} P_{\text{S}} + \frac{2}{3} P_{\text{NS}}$. The NS part is made up of spheroids with the same AR distribution as [Mishchenko et al., 1997].</td>
<td>Applying purely NS phase fns into satellite retrievals does not significantly improve them. Using both spherical and NS particles, a “great improvement” is found.</td>
</tr>
</tbody>
</table>
Table 2.5 continued . . .

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>Assumed properties</th>
<th>Conclusions</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Kalashnikova and Sokolik, 2004]</td>
<td>DDA and Mie. It would appear also T-matrix has been used (although not explicitly mentioned).</td>
<td>Four “representative” distributions containing in varying proportions spheres, spheroids, random shapes and hexagonal and rectangular disks. Spheroid distribution is as in Mishchenko et al. [1997].</td>
<td>i) Angular, sharp-edge particles cause largest differences in optical properties relative to spheres and spheroids increasing $\bar{\omega}_0$ and $\tau$; ii) Effect of NS on $\bar{\omega}_0$ is more important for strong absorptive material; iii) “Because sharp-edge particles are often a major fraction [of total aerosol], NS effect cannot be ignored”.</td>
</tr>
<tr>
<td>[Dubovik et al., 2006]</td>
<td>T-matrix [Mishchenko and Travis, 1998] &amp; ray tracing [Yang and Liou, 1996] for larger particles.</td>
<td>There are no particles with $AR &lt; 1.44$. The distribution of AR is a step function with $n(AR) = 0$ when $0.7 &lt; \epsilon &lt; 1.4$.</td>
<td>Stability of NS aerosol retrievals are increased if there is equal presence of prolate and oblate spheroids.</td>
</tr>
<tr>
<td>[Muñoz et al., 2007]</td>
<td>Ray tracing “Realistic shapes” from the Amsterdam database [Volten et al.].</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>[Yang et al., 2007]</td>
<td>As Dubovik et al. [2006]. For large particles, surface roughness is also tested. TOA radiances calculated.</td>
<td>Assumed that all particles are spheroids with $AR = 1.7$ (both oblate and prolate). Also reviewed [Nakajima et al., 1989, Okada et al., 1987, Hill et al., 1984, Reid et al., 2003] finding that “mode or mean AR were 1.7, 1.4, $\sim 2$, 1.9 and 2.2”.</td>
<td>“Non-sphericity effect of dust particles is significant at short wavelengths, however, not at the thermal infrared”.</td>
</tr>
</tbody>
</table>
Table 2.5 continued . . .

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>Assumed properties</th>
<th>Conclusions</th>
</tr>
</thead>
<tbody>
<tr>
<td>[Hudson et al.,</td>
<td>“Small particle regime”</td>
<td>Looked at needles, disks and continuous distributions of ellipsoids.</td>
<td>Disk simulations are the best fit for small clay particles.</td>
</tr>
<tr>
<td>2008]</td>
<td>[Mishra and Tripathi, 2008]</td>
<td>T-matrix Used spheres, oblate spheroids with $AR = 1.5$ and Chebyshev particles.</td>
<td>“Effect of [hematite] variation on $\tilde{\omega}_0$ is found to be stronger than particle nonsphericity”.</td>
</tr>
<tr>
<td>[Mishchenko and Travis, 1998]</td>
<td>T-matrix</td>
<td>Oblate and prolate spheroids with $\epsilon = 1.7$ as “this was the median value found from SEM analysis”.</td>
<td>-</td>
</tr>
</tbody>
</table>

From Table 2.5 we find that the most common method of modelling of mineral dust aerosol encountered was with Mishchenko and Travis’s T-matrix code [Mishchenko and Travis, 1998]. Spheroids were the most common NS shape used, one of two tactics for describing the shape distribution being employed. Some works picked a single representative aspect ratio, generally with a value of $\sim 1.6$ [Yang et al., 2007, Mishra and Tripathi, 2008, Osborne et al., 2008], while others used a distribution of aspect ratios with equal probability [Mishchenko et al., 1997, Wang et al., 2003, Kalashnikova and Sokolik, 2004, Dubovik et al., 2006]. Ray tracing was sometimes used for the more randomly shaped particles, or where the T-matrix method was not possible [Haywood et al., 2003, Dubovik et al., 2006, Yang et al., 2007]. DDA was not generally used in studies of this sort\textsuperscript{7}, since it places an unfeasibly large strain on computing resources in most cases. All studies conclude that including NS components of aerosol in light scattering models improve the agreement of phase functions with those observed. Interestingly, Wang et al. [2003] noticed that using only NS aerosol did not give so good an improvement in satellite retrievals as a mixture of spherical and NS particles.

Electron microscopy measurements were found to provide the most information on particle shape. The overall picture from Table 2.4 is of particles with rough edges and aspect ratios of $1.3 \rightarrow 2.2$. The AR distributions are generally reported to be independent of size, at least for all but the largest particles where Reid et al. [2003] found that particles with radius $> 10 \mu m$ were more elongated.

\textsuperscript{7}with the exception of Kalashnikova and Sokolik [2004]
In the case of Kandler et al. [2007], the distribution of aspect ratios, \( n(AR) \) was assumed to be a modified log-normal function given by,

\[
n(AR) = \frac{1}{\sqrt{2\pi\sigma(AR-1)}} \times \exp \left[ -\frac{1}{2} \left( \frac{\ln(AR-1) - \mu}{\sigma} \right)^2 \right],
\]

with \( \sigma = 0.6579 \) and \( \mu = -0.4502 \) giving a median aspect ratio \( AR = 1.65 \). The fit to this curve is very convincing (see Fig. 10 from Kandler et al. [2007]).

The larger particles were generally found to be more “angular” or have “sharper edges” than those which were smaller [Haywood et al., 2003, Reid et al., 2003, Muñoz et al., 2007, Chou et al., 2008], although this does not require the AR of a shape to change. Yang et al. [2007] used ray tracing to give larger particles a “rougner” surface. This was justified by inspection of electron micrographs of dust particles [Reid et al., 2003]. Another option with larger particles is to consider them as “irregular aggregates” with rougher contours [Reid et al., 2003, Chou et al., 2008]. This has not been used to model mineral dust in any of the papers reviewed.

Some sources also commented that a thin coating of sulphate (\( \sim 60\text{nm} \)) was found on samples [Kandler et al., 2007, Chou et al., 2008].

### 2.3.2 Method

As in the majority of previous studies, the T-matrix method is used to calculate the light scattering from spheroid mineral dust particles.

#### Different distributions

Fig. 2.11 shows four possible aspect ratio distributions we can use to give our best description of aerosol properties in the atmosphere. Our current assumption of sphericity is the yellow distribution. We also looked at other distributions in AR space, as used in previous studies.
Figure 2.12: Phase functions for spheroid AR distributions (as given in Fig. 2.11) with a log-normal distribution of particle size. Relative difference between the spheroid distributions, and a spherical-only distribution are shown in the right hand figure. Spheroid distributions use equal numbers of oblate and prolate spheroids.

- Polydisperse with equal probability of all ARs between 1.5 and 2.5.
- Monodisperse, with AR = 1.6.
- Polydisperse, with log-normal distribution, as given in [Kandler et al., 2007] and explicitly stated as (2.8).

Example phase functions for these aerosols, with log-normal size distributions are shown in Fig. 2.12. This case has a distribution effective radius of 2.26 \( \mu \text{m} \) (towards the larger end of mineral dust distributions). As usual, a lognormal distribution of sizes is used, and the only difference between the lines are the distributions of nonsphericity. The relative difference between the new, NS models and the spherical case are shown in the right hand plot. Initial impressions are that the phase function differences are very large in the back scatter, while manageable in the forward direction. Additionally, the differences from the spherical distribution are similar for the three NS distributions, that is, all three distributions alter the phase function in a similar way.

### 2.3.3 Results

Taking the most physically-justifiable distribution from Fig. 2.11, that of Kandler et al. [2007], we investigate the changes in phase function over a range of effective radii in Fig. 2.13. Most notably, with increasing particle size, the differences between the NS and spherical distributions become more pronounced. Biggest differences occur at scattering angles between 100° and 150° and are always positive, agreeing with previous findings by Mishchenko et al. [1997]. For the larger particles,
Figure 2.13: Relative difference between NS to spherical phase functions over a range of particle sizes. Distribution of NS particles is that of Kandler et al. [2007] as shown in Fig. 2.11.

Differences between the expected intensity at these angles can be as large as 200%. In the backscatter direction ($\theta > 150^\circ$), negative differences occur, but generally these are much less than the positive differences at slightly lesser scattering angles. For distributions with effective radii $< 1\mu m$, the mean relative difference in the phase function is less than 10%.

According to Dubovik et al. [2006], problems occurred when only oblate, or only prolate spheroids were used. It was reported that “the assumption of an equal presence […] resulted in improved stability of the retrievals”. Comparing these individual functions to the full function, Fig. 2.14 shows how oblate-only and prolate-only functions differ substantially from their combined function. The errors shown are mirror images of each other, as the full function is a combination in equal parts of the two other functions. Errors of over 10% in the backscatter direction for these plots present a serious worry. It is in no way clear whether oblate or prolate spheroids (or a combination of the two) are the most effective representations for mineral dust. That using one or the other will affect scattering properties so greatly (although not as greatly as with spherical comparisons) leads to an arbitrary choice in this exercise.

### 2.3.4 Applying the new model to retrievals

Look-up tables were created to be passed into ORAC retrievals over the Saharan area. Classes of aerosol are defined as a combination of several aerosol modes, each with their own refractive index,
### Table 2.6: Showing the variation in optical properties that a partially non-spherical mineral dust class causes.

<table>
<thead>
<tr>
<th>WL $[\mu m]$</th>
<th>$\beta_{ext} \times 10^{-5} [\text{km}^{-1}]$</th>
<th>$\langle \cos \theta \rangle$</th>
<th>$\bar{\omega}_0$</th>
<th>$P(180^\circ)$</th>
<th>$P(\text{max error})$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ORAC</td>
<td>NS</td>
<td>RE</td>
<td>ORAC</td>
<td>NS</td>
</tr>
<tr>
<td>0.55</td>
<td>3.32</td>
<td>3.33</td>
<td>0.38%</td>
<td>0.72</td>
<td>0.71</td>
</tr>
<tr>
<td>0.66</td>
<td>3.17</td>
<td>3.21</td>
<td>1.17%</td>
<td>0.71</td>
<td>0.71</td>
</tr>
<tr>
<td>0.863</td>
<td>3.00</td>
<td>3.07</td>
<td>2.04%</td>
<td>0.69</td>
<td>0.70</td>
</tr>
<tr>
<td>1.59</td>
<td>2.68</td>
<td>2.74</td>
<td>2.10%</td>
<td>0.69</td>
<td>0.69</td>
</tr>
<tr>
<td>10.86</td>
<td>0.96</td>
<td>0.96</td>
<td>0.61%</td>
<td>0.56</td>
<td>0.57</td>
</tr>
<tr>
<td>12.05</td>
<td>1.00</td>
<td>1.07</td>
<td>7.34%</td>
<td>0.55</td>
<td>0.55</td>
</tr>
</tbody>
</table>
median radius and radius standard deviation. In the mineral dust class (which currently consists of four aerosol modes), the spherical coarse, accumulation and nucleation components of mineral dust in the aerosol tables were replaced with spheroid minerals as shown in the previous section. The final component, a water soluble aerosol, was left spherical. The effective radius of the classes are calculated by modifying the mixing ratios of each aerosol mode so that the total effective radius matches that required.

**Scattering examples for the mineral dust class**

Fig. 2.15 shows a comparison of phase functions between a new version of the mineral dust aerosol class, incorporating non-spherical components, and the standard, spherical-aerosol-only version. Additionally, the DISORT radiative transfer model does not deal directly with phase functions, but instead, with Legendre coefficients\(^8\). Although this can lead to errors (if the series is terminated before high order terms are close enough to zero to make them negligible), results here show that errors are very small compared to the differences between spherical and NS aerosol classes. A mineral dust class effective radius of \(r_{\text{eff}} = 1.13 \mu m\) is shown, that size being chosen because it lies in the range of expected \(r_{\text{eff}}\) values of \(1 \rightarrow 2 \mu m\) [Hess et al., 1998].

As discussed previously, the change in aerosol shape has caused the phase function to increase in the backscatter direction, but decrease for scattering angles of \(90^\circ \rightarrow 150^\circ\). In the infrared channel shown, differences are much less, since the size parameter for longer wavelengths is, for the most part, less than unity\(^9\). Errors in the phase function at visible wavelengths peak at over 50 % in this case,

---

\(^8\) Expansion coefficients for the phase function in terms of Legendre polynomials [Arfken and Weber, 1995].

\(^9\) For \(\lambda = 12.05 \mu m\) and \(r_{\text{eff}} = 1.13 \mu m\) as in the current example, the effective size parameter is \(x_{\text{eff}} = 0.59\).
Figure 2.15: Mineral dust class phase functions for three selected AATSR channels. The effective radius in this case is \( r_{\text{eff}} = 1.13 \mu m \). NS and spherical models have been used and compared. In both cases, the refractive index, median radius \( (R_m) \), standard deviation of \( \ln r \) (S), and external mixing ratios of each aerosol component are kept constant. The exact calculations, and those recalculated from a terminated series of Legendre coefficients are shown.
at scattering angles of around 120°. This is remarkably convenient for AATSR whose sun-aerosol-
nadir viewing angle is generally in the range 110° → 130°. In the forward view, generally between
70° → 90°, errors are much smaller, suggesting that this view would be less susceptible to errors in
retrievals due to spherical assumptions.

Impressively, although we see large changes in the phase functions between the two versions of
mineral dust, the aerosol component, “Water Soluble, 50% humidity”, which is spherical, actually
accounts for 87% of the aerosol in the class at this radius. Additionally, of the sand components,
over 50% of the next most populous mode (the nucleation mode, with 11% of aerosol) have a size
parameter of less than 1. That leaves a very small proportion of particles (maybe 6%) which are
non-spherical, and are large enough to have phase functions which significantly deviate from the
spherical case.

Other effective radii (not shown) indicate the trend is as might be expected. For small particles,
the size parameters are smaller and when the wavelength of light becomes much greater than the
radius, the shape of the scattering particle becomes unimportant, and errors relative to the spherical
case decrease rapidly. Conversely, for larger particles, differences in shape become more pronounced,
and errors relative to the spherical case increase accordingly. Additionally, for the larger particles,
Legendre coefficients, terminated after 1000 terms (as is currently the case) fail to accurately repro-
duce the scattering patterns. Since the inclusion of NS particles tends to blur the phase functions,
making them less variable, this becomes less of a problem.

Top of atmosphere differences

After generating scattering properties for each aerosol class, the next stage is to put these properties
into a forward model which gives top of atmosphere reflectance. Fig. 2.16 shows relative differences
in reflectance between the non-spherical class ($R_{A36}$) and the spherical class ($R_{A03}$), as would be seen
by AASTR.

For the smaller effective radii (< 0.10µm), there is very little difference between the two models.
As particle size increases, the non-spherical particles are reflecting more light than their spherical
counterparts with differences of 30% at very large effective radii. This is not the only option, however.
Depending on the measuring angles of the satellite and the solar zenith angle, the changes can be
largely positive or largely negative. The scattering angles selected in this case have relevance to the
nadir view for AATSR in the example retrieval given in the following section.

Retrieval differences

Fig. 2.17 shows the results of an AATSR retrieval off the West African coast from the 10th March,
2006. A large dust plume can clearly be seen in the centre of the image.

The retrieval has found that, in general, within the dust plume, the NS aerosol class provides a
lower value of optical depth than the spherical aerosol. The effective radius is a less certain change,
with some sections of the plume having larger and some smaller.
Figure 2.16: Showing relative differences in the top of atmosphere reflectance arriving at satellite from a Lambertian forward model for the spherical and non-spherical mineral dust aerosol classes. This example is for a solar zenith angle of 60°, satellite zenith angle of 55° and a relative azimuth angle of 15°. The difference is shown as a function of aerosol effective radius and aerosol optical depth. A positive difference implies that the non-spherical aerosol has greater reflectance.

The cost difference\textsuperscript{10} shows that within the plume, the NS class sometimes has the lower cost, while at other positions, the spherical class wins out. This suggests that the NS class is not significantly better at predicting the scattering from sand than the current spherical model. The clearest difference in cost occurs over the area south west of the plume, where cost for the NS class is much higher. This is encouraging, as it suggests that the NS class is less likely to confuse non-mineral dust aerosol for mineral dust.

While these results provide a first glance at altering the scattering model for a known NS aerosol

\textsuperscript{10}The cost of a retrieval gives a measure of how much the measurements differ from the predicted forward model values, and how much the state we are inferring differs from the a priori information. In order to find our best estimate of a property, we minimise the cost function. The class is selected not only based on the class with minimum cost, but also due to constraints on the radiance and optical depth for the mineral dust class. For example, even if a mineral dust class has the lowest cost, if the optical depth is less than 0.4 it will not be selected.
Figure 2.17: Properties from AATSR retrievals of aerosol from 10th March, 2006. Location is over the Atlantic, just off the West African coast. The islands to the top right are part of the Azores archipelago. The false colour image provides a representation of the scene from space. The difference in optical depth, effective radius and cost are then shown. In all cases, the spherical result is subtracted from the non-spherical result.

in a retrieval, there is no way of knowing if they are representative without a more systematic study. Additionally, a particularly prominent dust storm was chosen as the first example. It would be of interest (although computationally expensive) to see how this new dust model alters the overall global statistics for optical depth and effective radius as well as cost.

2.4 Relative humidity

2.4.1 Background

Although areas where one expects mineral dust to be generated are dry, transport can be over thousands of miles and particularly across the Atlantic ocean. As such, one should expect to see mineral dust in a wide variety of humidity conditions. Fig. 2.18 shows average values and variability of relative humidity at surface level and confirms that an investigation of the scattering properties under changes in RH is warranted.
CHAPTER 2. IMPROVING THE OPTICAL REPRESENTATION OF SAND

2.4.2 Method

The mineral dust class was altered in an attempt to improve optical representation in a humid atmosphere. Several references help to build up a picture of sand composition during the SAMUM campaign. The smallest particles ($< 150$ nm) were seen to be the most hygroscopic (100\% soluble by volume), and are most likely ammonium sulphate [Kaaden et al., 2009]. It was also observed that above this size range was a “less hygroscopic” regime which was 40\% soluble by volume (assuming the solute was ammonium sulphate). At the upper range of the hygroscopic aerosol, it was noted that smaller particles ($< 0.35 \rightarrow 0.5 \mu m$) contained “a considerable volatile and hygroscopic fraction” but that the larger particles “were almost entirely non-volatile and showed no significant humidity related particle growth” [Petzold et al., 2009]. These observations were also shown in Weinzierl et al. [2009, Fig. 5].

Kaaden et al. [2009, Fig. 11] found that the backscatter coefficient measured by lidar at 532 nm had no change as a function of the atmospheric relative humidity.

With this information in mind, the four-mode distribution is altered as explained in Table 2.7.

Mode 1 - Fully hygroscopic

The fully hygroscopic mode relies on the OPAC values of refractive index and mode median radius of the water soluble mode as a function of relative humidity. These values are shown in Fig. 2.19. Mie theory is still used for the light scattering calculations since we are looking at a solution which will be spherical and which is small compared to the wavelength. Since $r_m$ changes depending on the humidity, the mode distribution changes slightly from that seen in Fig. 2.1. As one would expect, increased relative humidity causes larger particles, and refractive index tends towards the value for water as the concentration of the solute decreases.
CHAPTER 2. IMPROVING THE OPTICAL REPRESENTATION OF SAND

<table>
<thead>
<tr>
<th>Mode</th>
<th>Current model</th>
<th>Relative humidity adjusted model</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>OPAC “Water soluble” mode with 50 % RH at all times</td>
<td>OPAC “Water soluble” mode with varying RH. This also alters the median radius of the mode.</td>
</tr>
<tr>
<td>2</td>
<td>OPAC “Mineral dust nucleation” mode (insoluble)</td>
<td>40 % by volume ammonium sulphate, the rest insoluble mineral dust. Scattering modelled as coated spheres. The inner sphere is the insoluble fraction, the outer “coating” is the ammonium sulphate solution. The width of the coating is determined by the relative humidity. When the relative humidity is not sufficient to add a coating a particle, a single refractive index is chosen for a non-coated sphere.</td>
</tr>
<tr>
<td>3</td>
<td>OPAC “Mineral dust accumulation” mode (insoluble)</td>
<td>As mode 2.</td>
</tr>
<tr>
<td>4</td>
<td>OPAC “Mineral dust coarse” mode (insoluble)</td>
<td>OPAC “Mineral dust coarse” mode (insoluble)</td>
</tr>
</tbody>
</table>

Table 2.7: Comparing two representations of the desert dust aerosol class.

Modes 2 & 3 - Partially hygroscopic

For our partially-hygroscopic aerosol classes, we must use modifications of Köhler theory which apply for soluble particles with insoluble fractions [Pruppacher and Klett, 1997]:

\[
\frac{\epsilon_a}{\epsilon_{sat, w}} = \exp \left[ \frac{2M_w \sigma_{s/a}}{RT \rho_w a} - \frac{\nu \Phi_s \epsilon_v M_s \rho_s r_d^3}{M_s \rho_w (a^3 - r_d^3)} \right].
\]  

(2.9)

Eqn. 2.9 has a plethora of undefined terms:

- \( M_w \) - Molecular weight of water (= 18.0160 g mol\(^{-1}\)).
- \( M_s \) - Molecular weight of the soluble salt (= 132.14 g mol\(^{-1}\) for (NH\(_4\))\(_2\)SO\(_4\)).
- \( R \) - Universal gas constant (= 8.314 J mol\(^{-1}\) K\(^{-1}\)).
- \( a \) - Wet particle radius.
- \( r_d \) - Dry particle radius.
- \( \rho_w \) - Density of water (= 0.998 g cm\(^{-3}\) at 1 atm, 20 °C).
- \( \rho_s \) - Density of soluble salt fraction (= 1.130 g cm\(^{-3}\) for (NH\(_4\))\(_2\)SO\(_4\)).
- \( \epsilon_v \) - Water soluble volume fraction.
- \( \nu \) - Number of ions into which the salt will disassociate in water (3 for (NH\(_4\))\(_2\)SO\(_4\)).
- \( \sigma_{s/a} \) - Surface tension of solute / air boundary.
- \( \Phi_s \) - Molal osmotic coefficient of the salt in solution.

The majority of these values are easily obtainable, but two, \( \sigma_{s/a} \) and \( \Phi_s \) present some difficulty. Surface tension is a function of molal concentration of salt and temperature. From Pruppacher and
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Figure 2.19: Variation of OPAC water soluble aerosol mode properties with relative humidity. Median radius \( r_m \) is shown in the left hand plot, real part of refractive index \( n \) in the centre, and imaginary part \( k \) on the right. The values \( n \) and \( k \) are shown at the AATSR channel wavelengths.

Klett [1997], we obtain

\[
\sigma_{s/a}(m, T) = \sigma_{w/a}(T) + B m,
\]

(2.10)

where \( \sigma_{w/a}(T) \) is the temperature-dependant surface tension between a water/air boundary and \( m \) is the molal concentration. The gradient, \( B \), can be obtained from Jarvis and Scheiman [1967]. The molal osmotic coefficient, \( \Phi_s \), characterises the deviation of a solvent from ideal behaviour according to Raoult’s law, accounting for behaviour when the concentration of the solute is high. Since this is unlikely we set \( \Phi_s = 1 \).

We are interested in the relationship between \( a \) and relative humidity, \( \text{RH} = \frac{e_a}{e_{\text{sat}, w}} \) given \( r_d \). We rearrange (2.9) to give:

\[
\ln \left( \frac{e_a}{e_{\text{sat}, w}} \right) = \frac{M_w}{\rho_w} \left[ \frac{X}{a} - \frac{Y}{(a^3 - r_d^3)} \right],
\]

(2.11)

\[
Z a^4 - X a^3 + (Y - Z r_d^3) a + X r_d^3 = 0.
\]

(2.12)

where

\[
X = \frac{2\sigma_{s/a}}{RT}, \quad Y = \frac{\nu \Phi_s \epsilon_V \rho_i r_d^3}{M_s}, \quad \text{and} \quad Z = \frac{\rho_w}{M_w} \ln \left( \frac{e_a}{e_{\text{sat}, w}} \right).
\]

Although we can attempt to solve analytically for \( a \) in the quartic polynomial (2.12), underflow in the computations made this impossible, and the Newton-Raphson iterative technique was used to obtain our wet radius. Values of wet radius for an intial particle size of \( r_d = 0.1 \mu m \) as the relative humidity changes are shown in Fig. 2.20.

From the above discussion of field measurements, we chose \( \epsilon_V = 40\% \) [Kaaden et al., 2009]. The density of the insoluble mass fraction of mineral dust at the core was chosen to be \( \rho_i = 2.65 \text{ g cm}^{-3} \), which is representative of Saharan sand [Haywood et al., 2001].

After the wet radius has been calculated, further assumptions are required in order to calculate light scattering properties. Since the soluble portion of the mineral dust has now dissolved into
Figure 2.20: Showing how changing the relative humidity over an aqueous solution formed from an aerosol with soluble and insoluble fraction alters the drop radius. The initial size of the particles, $r_d = 0.1 \, \mu m$. The four curves show particles with different soluble fractions by volume, $\epsilon_V$. The upper half of the ordinate shows super-saturation and has a smaller scale.

solution, the volume of the sand particle core is reduced. To simplify calculations, we assume a coated sphere using a modification of $\texttt{bhcoat.f}$ written by Bohren and Huffman [1983] (which was converted to IDL and updated, so that the phase function is also calculated). Although there is available code to calculate light scattering by non-spherical inclusions to spherical scatterers [Doicu et al., 2006], it was felt that since the shape would be very poorly understood, computer overhead would be significantly greater, and that significant differences would be reduced over a large distribution of sizes, this was an unnecessary complication. Knowing $a, r_d$ and $\epsilon_V$, the interior radius, $r_{\text{core}}$, can be easily obtained, by assuming a water / salt solution would take up the same volume as pure water:

$$\text{Vol. occupied by insoluble part} = \frac{4}{3} \pi r_{\text{core}}^3 = r_d^3 (1 - \epsilon_V) \frac{4}{3} \pi,$$

$$\Rightarrow r_{\text{core}} = r_d \sqrt[3]{1 - \epsilon_V}.$$  \hspace{1cm} (2.13)

For the case of a dry particle, we calculate a single refractive index, weighted by the mass of the two types of scattering material, the (now dry) ammonium sulphate and the insoluble fraction:

$$m_{\text{dry}} = \frac{(1 - \epsilon_V) \rho_i m_i + \epsilon_V \rho_s m_{\text{dry} (NH_4)_2SO_4}}{(1 - \epsilon_V) \rho_i + \epsilon_V \rho_s}.$$ \hspace{1cm} (2.14)

Here, $m = n + ki$ are values of complex refractive index.
Table 2.8: Showing the spread of optical values caused by a normally distributed set of RH values with mean 50% and standard deviation 30%. The values are for sand with $r_{\text{eff}} = 1.13 \, \mu m$ which is 40% soluble.

Mode 4 - Hydrophobic

Since the fourth mode was deemed completely insoluble, it was left in an identical state to that of previous calculations.

Further required decisions on method

Since the intended application of this method was to improve observed aerosol, it was decided that the distributions should be log-normal for the wet radius (since we were observing the particles which were presumed to have a coating). This means that for conditions with heightened relative humidity, one would be observing smaller (dry) sand particles than for a dryer atmosphere with the same aerosol effective radius.

2.4.3 Results

The final 4 modes were then combined in an identical method to the standard ORAC routine [Thomas et al., 2005]. These are shown in Figures 2.21, 2.22, and 2.23 for various AATSR wavelengths. Additionally, Fig. 2.21 has overlaid data from Fig. 2.2.
Figure 2.21: Optical properties at $\lambda = 550$ nm. Overlaid crosses (+) are the measurements previously shown in Fig. 2.2.
Figure 2.22: Optical properties at $\lambda = 863$ nm. With the exception of $\lambda$, all other properties are identical to those of Fig. 2.21.
Figure 2.23: Optical properties at $\lambda = 1.59 \, \mu m$. With the exception of $\lambda$, all other properties are identical to those of Fig. 2.21.
2.5 Summary

Table 2.9 gives a summary of the work in this chapter. We see that variability in refractive index has only a minor effect on the optical properties in comparison to relative humidity and non-sphericity. This is in some ways misleading since although we found that the spread of RI from FAO soil map data was small, there were significant offsets in the values of both $n$ and $k$. Particularly, the smaller value of $k$ changes the absorption of the mineral dust significantly compared to current ORAC values as seen in Fig. 2.9.

Both relative humidity and non-sphericity have significant effects on the optical properties. The effects of varying RH are seen in the absorption-related properties, such as $\bar{\omega}$, since they essentially involve making the aerosol less absorbing by adding water to the particles. The effect of changing the shape of particles is most clearly seen in the phase function, which has huge changes in certain scattering directions.
 CHAPTER 2. IMPROVING THE OPTICAL REPRESENTATION OF SAND

Perturbation description & Variation caused to optical properties for $r_{\text{eff}} = 1.13\,\mu\text{m}$

<table>
<thead>
<tr>
<th>RI</th>
<th>Spread of values defined by FAO digital soil map of the world</th>
<th>$\beta^{\text{ext}}$</th>
<th>$\langle \cos \theta \rangle$</th>
<th>$\omega_0$</th>
<th>$P(180^\circ)$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>&lt; 1 % 1 % N/A</td>
<td>0.55 1.59 12.05</td>
<td>0.55 1.59 12.05</td>
<td>0.55 1</td>
<td>15 % 13 % N/A</td>
</tr>
<tr>
<td>RH</td>
<td>Small mineral dust particles are defined as being 40 % soluble and RH = 50 ± 30% and the particles are modelled as coated spheres.</td>
<td>30 % 26 % 15 %</td>
<td>2 % 1 % 3 %</td>
<td>2 % 1</td>
<td>6 %</td>
</tr>
<tr>
<td></td>
<td>&lt; 1 % 2 % 7 %</td>
<td>1 % 1 % 1 %</td>
<td>1 % 1 % 12 %</td>
<td>37 % 4</td>
<td>2 % 6 %</td>
</tr>
</tbody>
</table>

Table 2.9: Summary of the impact of various parameters on aerosol optical properties, extinction ($\beta^{\text{ext}}$), asymmetry parameter ($\langle \cos \theta \rangle$), single scatter albedo ($\omega_0$), and phase function at a scattering angle of $\theta = 180^\circ$, $P(180^\circ)$. 
Chapter 3

Further work on SPARCLE

SPARCLE is a detector designed to measure the size and refractive index of particles. The setup is a capillary tube which pumps single particles through a cavity containing a laser beam and two light detectors. The laser’s line of sight is across the path of passing particles so that light is scattered as they pass through the light beam, measuring the particles’ phase function.

In last year’s report, the description of a forward model for SPARCLE and optimisation was discussed. Since then, the design of has changed significantly, requiring a redefinition of the forward model which is outlined in this chapter. Although the model has been outlined, it has not yet been implemented.

3.1 What has changed

The original design for SPARCLE was for detectors arranged facing the light scattering target, so that the glass covering would be perpendicular to light scattered from the target. As a result, the detectors were defined by their angular position relative to the incoming laser light which gave the forward scatter direction, $\theta = 0^\circ$. It was found that when building the instrument, that a vacuum was only possible with a rectangular enclosure, requiring the light detectors to be orientated parallel to the incoming laser beam. Inspecting different areas of the phase function must then be done by moving the detector along, parallel to the beam as shown in Fig. 3.1.

Since the detectors are protected by a layer of glass, the Fresnel equations are required to calculate how much light of each polarization state is transmitted through this glass. This is the problem which is covered in the following work. As in the previous design, we have a photo multiplier tube (PMT), and a diode array (LDA). Since the PMT is a single detector, we can assume that any light which hits its glass cover and is transmitted through will be measured. The LDA is more of a challenge, since there are multiple detectors in a very small space, and a gap between the glass protective screen and the detecting elements, which has the potential to increase refraction. The additional problem of multiple reflections inside the glass screen has been neglected.
CHAPTER 3. FURTHER WORK ON SPARCLE

Figure 3.1: New model of SPARCLE. Detectors are fixed parallel to the incoming light, a distance \( d_i \) from the scattering source and are positioned to measure different parts of the phase function by moving a distance \( x_i \) along the side of the detector. The light scattering angle is \( \theta_i \) and the incident angle at the detector is \( \psi_i \).

Figure 3.2: The displacement of the light beam by the glass screen which protects the LDA. If we know the location of a detector element, for example, number 4 which is at \((x_4, d_4)\), the thickness of the glass, \( d_{\text{glass}} \), and the distance between the glass and the detector elements, \( d_{\text{air}} \), as well as the glass’ refractive index, we can calculate the scattering angle, \( \theta \) with which light left the scattering particle in order to meet the detector at this point. \( \psi_i \) is the incident angle of the incoming light in the plane of the scattering; \( \psi_t \) is the angle of the transmitted light and \( l \) is the perpendicular offset caused by the glass.
3.2 New mathematical description

3.2.1 Geometry

The first problem is discovering what is the corresponding original scattering angle, $\theta$, measured by a specific point on the detector. If the horizontal displacement, $x$, and the perpendicular distance of the detector to the beam, $d$, are known then it can be seen from Fig. 3.1 that $\tan \psi = x/d$ and

$$\theta = \frac{\pi}{2} - \psi. \quad (3.1)$$

(assuming that light that reaches the glass is detected).

For the PMT, this is about all that needs to be done, but for the LDA, calculating $\psi$ can be more complicated. Inspection of Fig. 3.2 leads, by geometry to:

$$\sin (\psi_i - \psi_t) = \frac{l}{d_{\text{glass}}/\cos(\psi_t)}, \quad (3.2)$$

$$\Rightarrow l = \frac{\sin (\psi_i - \psi_t)}{\cos(\psi_t)} d_{\text{glass}}, \quad (3.3)$$

where we are interested in $l$, the perpendicular deviation of the original beam, caused by the glass. Using Snell’s law, with refractive index $m$,

$$l = \sin \psi_i \left[ 1 - \frac{\cos \psi_i}{\sqrt{m^2 - \sin^2 \psi_i}} \right] d_{\text{glass}}. \quad (3.4)$$

One can relate $x$ to $l$ by further inspection of Fig. 3.2 which yields:

$$\left[ \frac{l}{\cos \psi_i} + x \right] = (d + d_{\text{glass}} + d_{\text{air}}) \tan \psi_i, \quad (3.5)$$

so substituting $l$ from (3.3), and defining $s = \sin \psi_i$, then

$$s \left[ 1 - \frac{\sqrt{1 - s^2}}{\sqrt{m^2 - s^2}} \right] d_{\text{glass}} \frac{1}{\sqrt{1 - s^2}} + x = (d + d_{\text{glass}} + d_{\text{air}}) \frac{s}{\sqrt{1 - s^2}}, \quad (3.6)$$

$$\frac{sd_{\text{glass}}}{\sqrt{m^2 - s^2}} + x = (d + d_{\text{air}}) \frac{s}{\sqrt{1 - s^2}}. \quad (3.7)$$

This can be solved numerically, but if $d_{\text{glass}} \ll (d_{\text{air}} + d)$, then this simplifies to $\tan \psi = x/d$ as before.

3.2.2 Basis vectors required for calculating transmission

A more serious issue is whether light rays will reach the detectors at all if they meet the protective glass at an angle greater than Brewster’s angle. The Fresnel equations [Bohren and Huffman, 1983] give transmission coefficients based on the electric waves travelling parallel and perpendicular to
the plane of scattering. From Mie theory, we have the two states in the plane of scattering defined by the incoming and scattering vector, which give us polarization states \( \hat{\mathbf{p}}_1 \) and \( \hat{\mathbf{p}}_2 \). In order to calculate transmission at the detector, we need to move to the orthogonal basis \( \hat{\mathbf{p}}_p \) and \( \hat{\mathbf{p}}_s \) which are in the plane of scattering at the detector, shown in Fig. 3.3. We also know the vector \( \mathbf{d}_i \) which is the scattered light vector at the point where it meets the plane of the detector. The state \( \hat{\mathbf{p}}_1 \) is perpendicular to the scattering plane, defined by the incoming light which travels along the x-axis and the scattered light which is along \( \mathbf{d}_i \). This means that we can define

\[
\hat{\mathbf{p}}_1 = \frac{\hat{\mathbf{e}}_i \times \mathbf{d}_i}{|\hat{\mathbf{e}}_i \times \mathbf{d}_i|}.
\]

The second state is perpendicular to the scattered light and the first state, so can be defined

\[
\hat{\mathbf{p}}_2 = \frac{\hat{\mathbf{p}}_1 \times \mathbf{d}_i}{|\hat{\mathbf{p}}_1 \times \mathbf{d}_i|}.
\]

The directions of the polarization states in the detector plane can be defined from the direction of the incoming light and the normal to the surface, \( \hat{\mathbf{n}}_s \). This can also be used to calculate the incident angle at the detector, \( \psi \).

\[
\hat{\mathbf{p}}_p = \frac{\hat{\mathbf{n}}_s \times \mathbf{d}_i}{|\hat{\mathbf{n}}_s \times \mathbf{d}_i|}, \quad \hat{\mathbf{p}}_s = \frac{\hat{\mathbf{p}}_p \times \mathbf{d}_i}{|\hat{\mathbf{p}}_p \times \mathbf{d}_i|}, \quad \cos \psi = \frac{\mathbf{d}_i \cdot \hat{\mathbf{n}}_s}{|\mathbf{d}_i|}.
\]

Now that the two sets of orthogonal polarization states have been defined, converting between the two is trivial, using the vector dot product.
Chapter 4

Time line and Future Work

4.1 Work to date

Michaelmas Term 2007

- Reacquainting with IDL and FORTRAN.
- Background reading on Mie theory and inverse methods.
- Familiarised myself with the EODG Mie routines\(^1\).
- Studied the AOPP 4th year undergraduate course and completed problem sets (this continued for the three terms).
- Began forward model for SPARCLE instrument.

Hilary Term 2008

- Extended the SPARCLE forward model, and wrote code to optimise the position of its detectors.
- Background reading in the T-matrix method.
- Attended workshop on non-spherical scattering in Bremen.
- Obtained Michael Mishchenko’s FORTRAN T-matrix code\(^2\) and modified, converting the main routines into callable procedures.

\(^1\)http://www-atm.physics.ox.ac.uk/code/mie/
\(^2\)http://www.giss.nasa.gov/~crmim/t_matrix.html
Trinity Term 2008

- Learnt to programme in C.
- Created IDL dynamically loadable modules (DLMs) to call Michael Mishchenko’s T-matrix code in IDL.
- Attended APPRAISE science meeting at Leeds University.

Michaelmas Term 2008

- Attended ELS-XI conference at University of Hertfordshire.
- Attended ADIENT meeting at University of Reading - Brief presentation.
- Attended NAG course on using the HeCTOR super computer.
- Attended ‘Introduction to Aerosol Science’ course at UCL.
- Further work on IDL DLMs, adding stability, generating look up tables for lengthy calculations of light scattering.
- Literature review on mineral dust aerosol.
- Work on SPARCLE

Hilary Term 2009

- Further work generating look up tables and integrating functions
- Created IDL version of Bohren and Huffman [1983] coated sphere code, bhcoat.f and added additional functionality (calculation of phase function and backscatter coefficient which are not included in the original code).
- ADIENT meeting at University of Manchester - Presentation on non-spherical scattering
- Submitted ADIENT report 4.1.2. - Assessing errors in optical properties due to particle asphericity and inhomogeneity.

Trinity Term 2009

- Investigation into the current state of the ORAC optical model sand.
- Work on relative humidity.
- Work on refractive index.
- Wrote IDL code to analyse FAO digital soil map data formats.
4.2 Plan for future work

4.2.1 Mineral dust aerosol

The main body of work on the mineral dust aerosol class has now been completed. Over the next year, various optical schemes described will be passed to ORAC mineral dust retrievals to see which appear best to improve the mineral dust optical properties.

Similar work will then be carried out on two additional aerosol types, trying as best as possible to mirror work on the mineral dust class.

4.2.2 Sea salt aerosol

Marine / salt aerosol is mainly affected by relative humidity. It would be hoped that RH calculations already carried out for the mineral dust class will help speed up the implementation of similar calculations for this aerosol class.

Other serious issues with this class include non-sphericity when dry, and deliquescence / efflorescence hysteresis curve when the relative humidity changes. Size distribution and refractive index will also be investigated.

4.2.3 Aerosol from burning

As mentioned in Chapter 1, burning of aerosol creates all kinds of horrible problems for modellers. These include coagulation, fractal shapes, ageing and very poorly understood refractive index. As many of these subjects will be investigated as is possible in the remaining available time.
Bibliography


Craig F. Bohren and Donald R. Huffman. *Absorption and Scattering of Light by Small Particles*. Wiley-VCH, 1983. Chapter 4 has a very well explained derivation of Mie theory.


Cedric Chou, Paola Formenti, Michel Maille, Patrick Ausset, Guenter Helas, Mark Harrison, and Simon Osborne. Size distribution, shape, and composition of mineral dust aerosols collected during


E. J. Highwood. Suggested refractive indices and aerosol size parameters for use in radiative effect calculations and satellite retrievals. ADIENT / APPRAISE CP2 technical report, Department of Meteorology, University of Reading, August 2009. 1st Draft.


BIBLIOGRAPHY


