



Single scattering albedo of homogeneous, spherical particles in the transition regime

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ABSTRACT

Aerosol single scattering albedo (SSA) is the most important intensive particle parameter controlling aerosol direct radiative forcing. For homogeneous, spherical particles and a complex refractive index independent of wavelength, the SSA is solely dependent on size parameter (ratio of particle circumference and wavelength) and complex refractive index of the particle and can be accurately calculated with Mie theory. Here, we explore this dependency for particles of intermediate size in the transition or peak regime between the Rayleigh scattering and geometric optics regimes. We show that in the transition regime, low-frequency oscillations (the interference structure) of SSA as function of size parameter occur for relatively small imaginary parts of the refractive index and are caused by interference between transmitted and diffracted components of the scattered light. Anomalous diffraction theory (ADT) semi-quantitatively describes this behavior of the SSA as function of size parameter and complex refractive index. While ADT accurately gives the size parameters of the interference peaks, ADT amplitudes only approximate exact Mie results. A significant improvement in agreement with Mie theory can be obtained with modified ADT (MADT) that adds a parameterization for the physics neglected in ADT, namely internal reflection/refraction, photon tunneling, and edge diffraction.

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1. Introduction

The aerosol single scattering albedo (SSA) is the most important intensive particle parameter controlling aerosol direct radiative forcing. A simple analytical equation given by Chýlek and Wong [5] estimates clear-sky aerosol radiative forcing as function of SSA and aerosol upscatter fraction. To use this equation the aerosol upscatter fraction must be related to intensive aerosol properties that can be measured or calculated, that is to the aerosol backscatter fraction or asymmetry parameter [17]. Recently, the aerosol radiative forcing equation of Chýlek and Wong [5] has been evaluated by comparing its estimations with the output of a global Monte-Carlo Aerosol Cloud Radiation (MACR) model; its estimations have been found adequate for clear-sky conditions [7].

The SSA, the ratio of scattering and extinction efficiencies, Q_{sca} and Q_{ext} , respectively (where efficiencies Q are the ratio of optical and geometrical cross-sections and the extinction efficiency is the

sum of scattering and absorption efficiencies, $Q_{ext} = Q_{sca} + Q_{abs}$) can be written as

$$SSA = \frac{Q_{sca}}{Q_{ext}} = \frac{Q_{sca}}{Q_{sca} + Q_{abs}} = \left[1 + \frac{Q_{abs}}{Q_{sca}} \right]^{-1} = 1 - \frac{Q_{abs}}{Q_{ext}}, \quad (1)$$

where $0 \leq SSA \leq 1$; 0 for purely black, non-scattering but absorbing particles and 1 for purely white, scattering but non-absorbing particles.

For homogeneous, spherical particles, the SSA can be obtained easily from Mie theory [9] calculations as function of particle complex refractive index $m = n + i\kappa$ and particle size parameter $x = \pi D/\lambda$, where n and κ are the real and imaginary parts of the particle refractive index, respectively, D is the particle diameter, and λ the wavelength of the incident light. Note that the bulk or material absorption coefficient $\alpha = 4\pi\kappa/\lambda$ is directly related to the imaginary parts of the refractive index. For a complex refractive index independent of wavelength, the SSA is solely dependent on size parameter x and complex refractive index m [11].

We build upon initial discussions of aerosol SSA size dependence by Moosmüller and Arnott [15], Moosmüller et al. [16], and Sorensen [21] to complement a recent, detailed discussion of SSA

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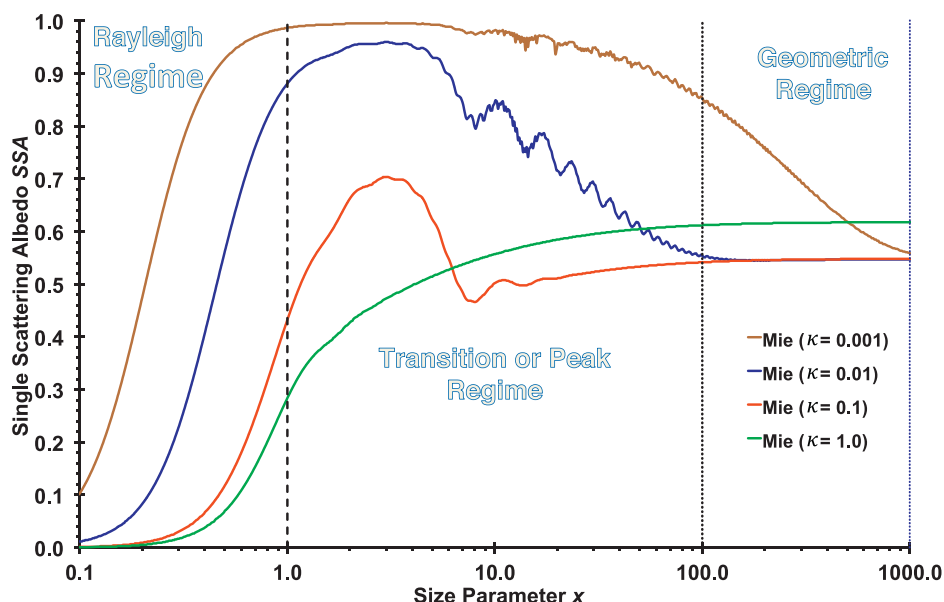


Fig. 1. Mie calculations of single scattering albedo SSA as function of size parameter x for a refractive index $m = 1.5 + i\kappa$. The vertical dashed line indicates the approximate boundary between Rayleigh and transition or peak regime, the vertical dotted black line the “generic” boundary between transition and geometric regimes ($x = 100$), and the vertical dotted blue line the same boundary for $\kappa = 0.001$, corresponding to $\kappa x = 1$.

for homogeneous, spherical particles in the small and large particle limits [18]. Here, we add a discussion of the SSA in the transition or peak regime between the small and large particle limits, where the SSA transitions from its x^3 dependency in the small particle limit (i.e., Rayleigh regime) to its constant value in the large particle limit (geometric optics regime) and has one or more peaks if the imaginary part of the refractive index is small enough. In Fig. 1, we show an overview of SSA calculated with Mie theory as function of size parameter x for several different values of the complex particle refractive index m , defined as

$$m = n + i\kappa, \quad (2)$$

where n is the real part and κ the imaginary part of the particle refractive index. In Fig. 1, we use a typical real part (i.e., $n = 1.5$) and vary the imaginary part κ over three orders of magnitude (i.e., from 0.001 to 1.0). In this figure, three different regimes can be distinguished and are approximately separated by vertical dashed lines, note however, that these boundaries depend on the complex refractive index and are not precisely defined: (1) The Rayleigh regime where $x \ll 1$ and consequently the incident light wave uniformly penetrates the particle and light scattered by the different sub-volumes of the particle is in phase, with amplitudes coherently adding. This leads to scattering and absorption cross-sections proportional to particle volume squared and volume, respectively [15], and to an SSA quickly increasing with size parameter x , proportionally to x^3 [18]; (2) The transition or peak regime, where SSA transitions from the its x^3 dependence in the Rayleigh regime to being independent of size parameter x in the geometric optics regime and where for small imaginary parts of the refractive index (i.e., $\kappa \ll 1/x$), the SSA shows peaks and ripples; and (3) the geometric optics regime of our everyday visual experience, where the SSA is independent of size parameter x and particle optics can be accurately calculated using geometric optics plus diffraction [14,18]. Note that for $\kappa \ll 1$, the boundary of the geometric optics regime can be estimated as $x\kappa = 1$; Fig. 1 shows a “generic” boundary of $x = 100$ and separately, for $\kappa = 0.001$, the appropriate boundary of $x = 1000$, corresponding to $x\kappa = 1$. Therefore, the boundaries of the SSA Rayleigh, transition, and geometric optics regimes depend on the imaginary part of the refractive index κ ; for example for large κ (i.e., $\kappa = 0.1, 1.0$) SSA is nearly constant for size parameter

$x > 100$, indicating the beginning of the geometric optics regime, while for small κ (i.e., $\kappa = 0.001$), this does not happen until the product (κx) becomes larger than 1. The dependence of regime boundaries on the complex refractive index still needs to be investigated in more detail.

Recently, the small and large particle limits (i.e., Rayleigh and geometric optics regimes) of SSA have been discussed in detail [18] and here, we discuss SSA in the transition regime in the context of Fig. 1.

2. Single scattering albedo (SSA) in the transition regime

In the transition or peak regime, the SSA transitions from its x^3 proportionality in the Rayleigh (or small particle) regime to being independent of size parameter x in the geometric optics (or large particle) regime and for small imaginary parts of the refractive index (i.e., $\kappa \ll 1$), the SSA shows peaks and ripples superimposed upon these peaks. This behavior is more complex than that in the small or large particle limit, where SSA is determined by the Rayleigh scattering equation containing the Lorentz-Lorenz formula and the geometric optics equations, respectively [18]. Specifically, we want to know what physical phenomena cause the peaks and ripples. To this end, anomalous diffraction theory (ADT) can give us a physical, semi-quantitative understanding of the dominant SSA features in the transition regime, and its use will be discussed in the following section. More recently, modified anomalous diffraction theory (MADT) has been used as an improved parameterization for intermediate size particle optics [1,12], and we will explore the application of MADT to understand the transition regime behavior of SSA more quantitatively.

2.1. Anomalous diffraction theory (ADT)

In the transition regime we use the anomalous diffraction theory (ADT) of van de Hulst [22] for a simpler and more understandable description of particle scattering and absorption than that given by Mie theory. An alternative formulation of ADT has been given by Bryant and Latimer [4] and has been used, for example, by Mitchell [12] for his formulation of modified ADT (MADT). Quantitative results from the equations of van de Hulst [22] and

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