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Small and large particle limits of single scattering albedo for homogeneous, spherical particles

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ABSTRACT

The aerosol single scattering albedo (SSA) is the dominant intensive particle parameter determining aerosols 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. Here, we explore this dependency for the small and large particle limits with size parameters much smaller and much larger than one.

We show that in the small particle limit of Rayleigh scattering, a novel, generalized size parameter can be introduced that unifies the SSA dependence on particle size parameter independent of complex refractive index. In the large particle limit, SSA decreases with increasing product of imaginary part of the refractive index and size parameter, another generalized parameter, until this product becomes about one, then stays fairly constant until the imaginary part of the refractive index becomes comparable with the real part minus one. Beyond this point, particles start to acquire metallic character and SSA quickly increases with the imaginary part of the refractive index and approaches one.

1. Introduction

The aerosol single scattering albedo (SSA) is the dominant intensive particle parameter determining aerosol direct radiative forcing. Chýlek and Wong [6] have given a simple analytical equation that estimates aerosol radiative forcing as function of SSA and the aerosol upscatter fraction. This equation becomes very useful when the aerosol upscatter fraction is properly related to the aerosol backscatter fraction or the asymmetry parameter [17]. Recently, the aerosol radiative forcing equation of Chýlek and Wong [6] has been compared with the output of a global Monte-Carlo Aerosol Cloud Radiation (MACR) model and been found adequate for cloud-free conditions [8].

For homogeneous, spherical particles, the SSA can easily be obtained from Mie theory [13] calculations as function of the particle complex refractive index m with

$$m = n + i\,\kappa, \tag{1}$$

where n and κ are the real and imaginary parts of the refractive index, respectively, and the size parameter x with

$$x = \frac{\pi D}{\lambda}, \tag{2}$$

where 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. We build upon the initial discussions of aerosol SSA size dependence by Moosmüller and Arnott [15], Moosmüller et al. [16], and Sorensen [19]. In Fig. 1, we show a log-log plot of single scattering albedo (*SSA*) calculated with Mie (solid lines) and Rayleigh (dashed narrow lines) theory as function of size parameter x for several different values of the imaginary part κ of the particle refractive index and a typical real part (i.e., $n = 1.5$). In this figure, three different regimes can be distinguished and are approximately separated by vertical dashed lines: (1) The Rayleigh regime where $x << 1$ 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,

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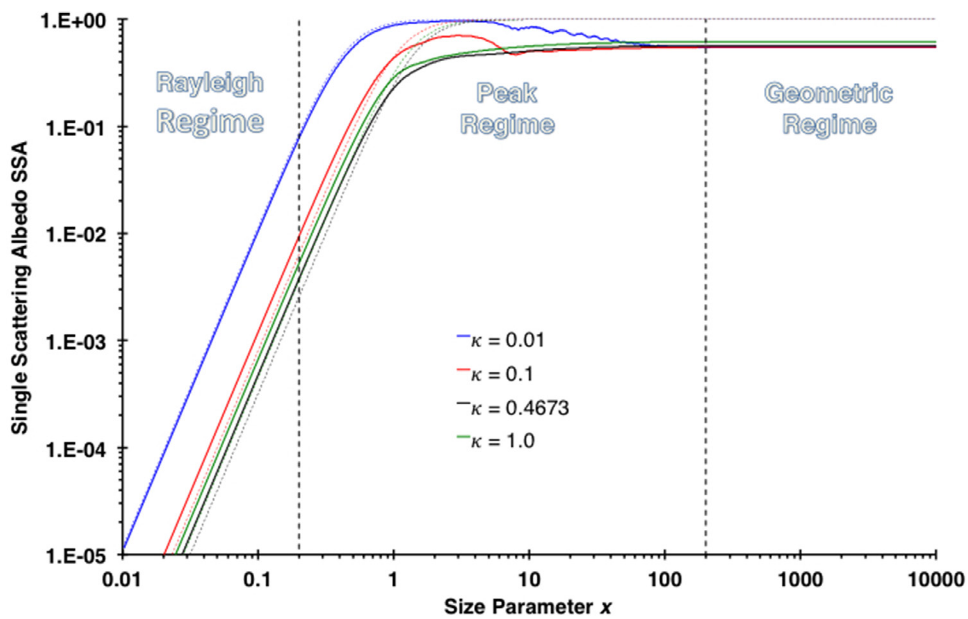

Fig. 1. Single scattering albedo SSA as function of size parameter x for a refractive index $m = 1.5 + \mathrm{i}\ \kappa$ with Mie and Rayleigh calculations shown as solid and dashed lines, respectively. Vertical dashed lines indicate approximate boundaries between Rayleigh, peak, and geometric regimes. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

respectively, and to an SSA quickly increasing with size parameter x; (2) The "peak" regime, where for small imaginary parts of the refractive index (i.e., $\kappa << 1$), the SSA peaks and shows ripples; and (3) the geometric optics regime of our everyday visual experience, where the SSA is independent of size parameter x. In the following, we will discuss the small and large particle limits of SSA in the context of Fig. 1.

2. Small particle limit

In the small particle limit ($x << 1$) we can use Rayleigh theory for a simpler and more understandable description of particle scattering and absorption than given by Mie theory [4]. Rayleigh theory uses the Lorentz-Lorenz factor $LL(m)$ [11,12] given by

$$LL(m) = \frac{m^2 - 1}{m^2 + 2}, \tag{3a}$$

with $E(m)$ and $F(m)$ conventionally used to denote imaginary part and complex square of LL, respectively [19] as

$$E(m) = Im\{LL(m)\} \ and \ F(m) = |LL(m)|^2. \tag{3b}$$

In the Rayleigh regime ($x << 1$), Rayleigh particle scattering efficiency Q_{sca_Ray} and absorption efficiency Q_{abs_Ray} (both ratios of optical to geometric cross-section) can be written simply as

$$Q_{sca\_Ray}(x, m) = \frac{8}{3} x^4 F(m) \text{ and} \tag{4a}$$

$$Q_{abs\_Ray}(x, m) = 4xE(m). \tag{4b}$$

The SSA, the ratio of scattering and extinction cross-sections (where the extinction cross-section is the sum of scattering and absorption cross-sections) can be written in terms of efficiencies as

$$SSA(x, m) = \frac{Q_{sca}}{Q_{ext}} = \frac{Q_{sca}}{Q_{sca} + Q_{abs}} = \left[1 + \frac{Q_{abs}}{Q_{sca}}\right]^{-1}, \tag{5a}$$

where Q_{ext} is the extinction efficiency. Using the explicit expressions for Q_{sca_Ray} and Q_{abs_Ray} given in Eq. (4) yields SSA_{Ray}, the SSA in the Rayleigh regime as

$$SSA_{Ray}(x, m) = \left[1 + \frac{1.5}{\left(\frac{F(m)}{E(m)}\right) x^3}\right]^{-1}. \tag{5b}$$

Defining a function f of the complex refractive m as the ratio of $F(m)$ and $E(m)$

$$f(m) = \frac{F(m)}{E(m)} \tag{5c}$$

allows one to write

$$SSA_{Ray}(x, m) = \left[1 + \frac{1.5}{f(m)\ x^3}\right]^{-1}, \tag{5d}$$

where the dependence of SSA_{Ray} on size parameter x and refractive index m is cleanly separated. This expression can be expanded into a power series with respect to size parameter x as

$$SSA_{Ray}(x, m) = \frac{2}{3} f(m) x^3 - \frac{4}{9} f(m)^2 x^6 + \frac{8}{27} f(m)^3 x^9 - \ldots, \tag{5e}$$

where for $x << 1$, SSA_{Ray} is proportional to x^3 and to $f(m)$. With increasing x, higher order terms come into play and SSA_{Ray} converges monotonically to one (Fig. 1). However, as x increases, we leave the Rayleigh regime and Eq. (5) are no longer valid; the Rayleigh solution diverges from the Mie result (Fig. 1).

Within the Rayleigh regime, the refractive index function $f(m)$ determines the dependence of the SSA_{Ray} on the refractive index m. For a common real part of the refractive index, that is $n = 1.5$, $f(m)$ and therefore SSA_{Ray} have one minimum (i.e., local and global; see Fig. 2) for $\kappa_{min}(n = 1.5) = 0.4673$ with

$$f_{min}(1.5 + i\ \kappa) = f(1.5 + i\ 0.4673) = 0.7204 \text{ and consequently} \tag{6a}$$

$$SSA_{Ray\_min}(1.5 + i\ \kappa) = SSA_{Ray}(1.5 + i\ 0.4673) = \left[1 + \frac{1.5}{0.7204 x^3}\right]^{-1}. \tag{6b}$$

A plot of this minimum SSA_{Ray} as function of x for $\kappa = 0.4673$ is included in Fig. 1.

Obviously, the imaginary part of the refractive index yielding minimum $f(m)$ for any size parameter in the Rayleigh regime is a function of the real part n of the refractive index. Fig. 3 shows a contour plot of the refractive index function $f(m)$ as function of