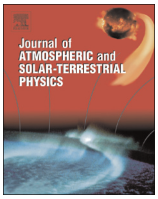
Journal of Atmospheric and Solar-Terrestrial Physics 133 (2015) 121–128

Contents lists available at ScienceDirect

Journal of Atmospheric and Solar-Terrestrial Physics

journal homepage: www.elsevier.com/locate/jastp



A model for absorption of solar radiation by mineral dust within liquid cloud drops

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ARTICLE INFO

Article history:
Received 13 April 2015
Received in revised form
25 August 2015
Accepted 26 August 2015
Available online 28 August 2015

Keywords:
Mineral dust aerosol
Light absorption
Cloud physics
Saharan air layer

ABSTRACT

Models of light scattering and absorption that consider the effect of insoluble inclusions present within liquid cloud droplets may assume the inclusion occupies random locations within the droplet. In certain cases, external forces can lead to certain orientations or alignments that are strongly preferred. Within this modeling study, we consider one such case in which an insoluble mineral dust inclusion ($\rho=2.6$ g/cm^3) is placed within a liquid water drop ($\rho=1.0$ g/cm^3). Such an instance mimics mineral dust aerosols being incorporated within cloud drops in Earth's atmosphere. Model results suggest super-micron mineral dust settles to the bottom of cloud droplets. However, Brownian motion largely randomizes the position of sub-micron mineral dust within the droplet. The inherent organization of the particles that result has important consequences for light absorption by mineral dust when present within a cloud drop. Modeled results suggest light absorption efficiency may be enhanced by as much as 4–6 fold for an isolated droplet experiencing direct solar illumination at solar zenith angles of $<20°$. For such an isolated droplet, the absorption efficiency enhancement falls rapidly with increasing solar zenith angle indicating a strong angle of incidence dependence. We also consider the more common case of droplets that contain dust inclusions deep within optically dense clouds. Absorption efficiency enhancements for these locales follow a dramatically different pattern compared to the optically isolated droplet due to the presence of diffuse rather than direct solar irradiation. In such cases, light absorption efficiency is decreased through including super-micron dust within water droplets. The study has important implications for modeling the absorption of sunlight by mineral dust aerosol within liquid water clouds. The angle of incidence dependence also reveals that experimental measurement of light absorption for cases in which particle alignment occurs may not always accurately reflect atmospheric absorption of sunlight. Therefore, care must be taken to extrapolate measurement data to climate models.

1. Introduction

Mineral dust particles represent one of the largest mass fluxes of any particle type into Earth's atmosphere as estimated as 1–3 Pg yr^{-1} (Hinds, 2012; Redmond et al., 2010; Textor et al., 2006). While the dust itself can alter visibility dramatically by absorbing and scattering sunlight and affect human health, it can also influence the properties of clouds and weather patterns on an episodic basis. For instance, Min et al. (2009) have found evidence that mineral dust may suppress precipitation in certain cases. Rosenfeld et al. (2001) also suggests Saharan dust suppresses precipitation and cloud droplet growth through a mechanism that slows coalescence of droplets. Models of different air masses suggest mineral dust has a lesser effect on precipitation (Teller et al., 2012; Yin et al., 2002; Zhao et al., 2011). However, the presence of the hot, dry, and dusty Saharan air layer is known to suppress cumulus convection and may have some influence on the development of tropical cyclones through either heating the airmass aloft thru absorption of solar radiation, cooling ocean surfaces, or both (Braun et al., 2013; Carlson and Prospero, 1972; Dunion and Velden, 2004; Sippel et al., 2011).

The presence of insoluble mineral dust inclusions within cloud drops appears to be a common occurrence. Matsuki et al. (2010) found that a majority fraction of cloud drop residuals collected over Niger contain evidence of mineral dust. In a recent paper, Twohy (2015) reports that approx. 1/3 of cloud drops within a convective cloud over the eastern tropical Atlantic contain mineral dust. In addition, Cziczo et al. (2013) and Targino et al. (2006) have demonstrated mineral dust is the dominant ice nuclei in cirrus clouds for a variety of locations.

Since mineral dust aerosol is known to be present within cloud drops, and the dust aerosol is known to absorb visible and infrared radiation, we have pursued the development of a model that

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http://dx.doi.org/10.1016/j.jastp.2015.08.013
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describes optical absorption by dust within liquid cloud droplets. The internal mixing of dust within cloud water drops may alter the visible optical properties of mineral dust aerosol. Therefore, such a modeling study is warranted. The data we report may motivate changes to weather or climate models to more accurately describe the absorption of solar light by mineral dust within or near liquid clouds.

The study of airborne hydrometeors containing absorptive inclusions is not a new research topic. Extensive literature has contributed knowledge for hydrometeor mixing with various materials. Among them, black carbon (BC) is regarded as the most significant absorptive agent. Martins et al. (1998) studied how the size and morphology of black carbon will contribute to light absorption. Chylek and Hallett (1992) and Chylek et al. (1985) presented experimental evidence suggesting that black carbon remains at the surface of water drops, and modeled a strong electric field-enhancement that occurs leading to very high light absorption. Such absorption enhancement would contribute to enhanced atmospheric heating which can't be neglected in models of aerosol absorption. Jacobson (2001, 2012) has long championed the idea that when absorptive materials exist as an internal mixture, the light absorption can be increased and has studied the climate consequences of this added absorption. However, upon close examination of the 1985 and 1992 works by Chylek et al., the data presented reflects specific mode resonances or the discussion includes locations along the main optical axis of the drop studied. Both of these cases would yield large or very large modeled absorption enhancements, and it is not clear to us that the values reported are representative of surface positions of the droplet at a much more common non-resonant condition.

Nonetheless, Schnaiter et al. (2005), Wei et al. (2013), Shiraiwa et al. (2010), Khalizov et al. (2009) have all found experimental evidence for absorption enhancement in reasonably good agreement with Mie theory for core-shell type coating of the absorptive inclusion (Bond et al., 2006) for particles with outer diameters <2 μm. However, less compelling evidence exists for authentic atmospheric cases or cases in which the coating material is less volatile and/or a solid phase (Cappa et al., 2012; Knox et al., 2009; Lan et al., 2013; Thompson et al., 2012, Wei et al., 2013). The physical position of the absorptive inclusion within the dielectric is crucial to describing absorption enhancements.

Other absorptive aerosols such as mineral dust may also experience enhancement in absorption when mixed with other dielectric materials. Lack et al. (2009) has reported photoacoustic absorption measurements that suggest as humidity changed, the light absorption will be affected because of the water uptake by the aerosol. Results suggested an increase of absorption on the order of at least 1.5-fold in limited experiments. Due to complications of chemical and physical interaction of dust and environment, simulations of mineral dust's atmospheric light absorption remains a challenging topic (Sokolik et al., 2001).

Within this modeling study, we consider mineral dust as an inclusion ($\rho=2.6$ g/cm^3) placed within a liquid water drop ($\rho=1.0$ g/cm^3). Electric field intensity distributions based on a Mie scattering model are used to predict absorption enhancement for simulation purposes. A major assumption of the method is that the electric field distribution modeled within a droplet is a proxy of light absorption by an inclusion. This assumption may not be valid for inclusions that are strongly light absorbing since their presence within the droplet (and location) will affect the electric field. The current study considers cloud water drops >5 μm in diameter. This stands in contrast with the previously cited reports of absorption enhancements in internally mixed particles that are generally <2 μm in diameter. In addition, light absorption by dust particles at surface positions of the droplet are rigorously considered.

2. Methods

2.1. Mie scatter model

A spherical dielectric cloud droplet is studied in this work. The normalized electric field intensity ($|E|^2$) within the particle was determined by a Mie code (BHFIELD) developed by Suzuki and Lee (2008). The 'normalized electric field' (a.k.a. source function) we report is normalized to the incident wave having a value equal to 1. As a result, electric fields >1 represent an absorption efficiency enhancement and fields <1 represent a decrease in absorption efficiency. It is important to realize that the normalized field does not represent the absolute quantity of light that is absorbed by a dust grain, but rather the efficiency with which it absorbs. To locate the field intensity at certain place in the 3-D space, the spherical droplet is embedded in a 3-D computational box. The dimensions of grids in this cubic coordinate system can be adjusted through input commands. In addition, parameters including the wavelength of the incident light, size of the particle, and refractive indexes can be controlled by the user. For BHFIELD calculations, the droplet is modeled as illuminated by a monochromatic, plane polarized (parallel) wave propagating in $+z$ direction of the coordinate system with unit amplitude ($E_0=1$). This polarization was used exclusively, but deemed reliable due to the radial symmetry of the spherical droplet/inclusion. A basic assumption to be made for this code is that the whole particle and the medium that contains this particle should be homogeneous and isotropic. Birefringence of materials was not considered. The spherical water drop was modeled as a dielectric sphere of $m=1.33$. Internal electric fields within the water drop were determined without the presence of the dust inclusion as this was a requirement of the Mie code used. The Mie approach using BHFIELD allowed us to map the normalized electric field at a many coordinates within the water droplet. On occasion a particle size was employed that led to strong Mie resonance that increased the field dramatically. Results appeared as outlier data and were abandoned since this special, resonant case (of m, λ and diam.) is not representative of common atmospheric conditions. We direct the reader to Chylek et al. (1985) for a thorough discussion of absorption enhancement on resonance modes.

Discrete dipole approximation modeling of the droplet/dust particle was originally attempted, but quickly abandoned due to computational requirements that exceed our facility's capability for droplets with diameters >5 μm. Since internal fields were computed in the absence of the dust inclusion, and a central hypothesis of this manuscript is the proportionality of the electric field to light absorption, it became necessary to test this assumption. To accomplish this, we used the discrete dipole code (ADDA) (Yurkin and Hoekstra, 2011) to test the hypothesis that light absorption scales with electric field for smaller particles (<3 μm). For this experiment, identical droplets were considered in both BHFIELD and ADDA. In the ADDA runs, a dust inclusion was placed within the water drop at differing z-coordinates (along axis of light propagation) and the absorption cross section and Mie absorption efficiency term computed for each test particle. Then, BHFIELD was used to compute internal electric fields at identical conditions of diameter, λ, and m, but with no dust inclusion present. A refractive index of $m=1.53+0.0057i$ was used for dust inclusion at the wavelength of 530 nm in the ADDA calculations. The mean normalized electric field within the volume bounded by the hypothetical dust inclusion was then extracted from the data. Finally, the Mie absorption efficiency (Q_{abs}) was directly compared to the mean electric field experienced within the volume of the droplet containing the hypothetical dust inclusion as returned by BHFIELD. For ADDA simulations, both parallel and perpendicular plane polarizations are automatically averaged.