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A regional, size-dependent, and causal effective medium model for Asian and Saharan mineral dust refractive index spectra

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ABSTRACT

A model is developed for the refractive index spectra of desert mineral dust. This model is applicable regionally to both Asian and Saharan dust as the largest global aerosol sources. The capability of the model further aims at representing important local features through a subdivision into northern and southern Sahara, as well as western and eastern Asia. Machine learning techniques for accelerated literature data acquisition are presented. Available refractive index spectra for individual minerals and chemical species are combined based on the Bruggeman effective medium formula. A numerical procedure for effectively solving the resulting higher-order polynomial expression is presented. The present results of the effective refractive indices are validated through the Kramers-Kronig relation; in particular, a Hilbert transform is applied to the imaginary part of the refractive index spectra.

1. Introduction

The optical properties of terrestrial mineral dust have been a long-standing topic of investigation as evidenced by e.g. the investigations of Longtin, Shettle, Hummel, and Pryce (1988) or Carlson and Benjamin (1980) as well as Patterson, Gillete, and Stockton (1977), to name a few. During the last four decades, the interest of the respective contemporary investigations has gradually shifted from remote mineral exploration, as discussed by Egon and Hilgeman (1979) to the impact of mineral dust on the planetary radiation budget due to its relevance for the global climate and solar energy conversion in the Sahara. Similar to many scattering particles, mineral dust may reduce the radiative forcing by scattering shortwave radiation back into space, while on the other hand absorption of long wave radiation by absorbing constituents such as Hematite and soot may simultaneously have an opposite effect. The prediction of this balance between absorption and reflection giving rise to atmospheric warming and cooling respectively is still subjected to significant uncertainties. Relevant studies on the climate impact were performed by e.g. Shell and Somerville (2007) or Yue, Wang, Wang, and Fan (2009) while Schroedter-Homscheidt, Oumbe, Benedetti, and Morcrette (2013) and Polo and Estalayo (2015) discussed the improvement of solar power production by means of adequate aerosol forecasts and specific energy conversion process adaptions.

While the Sahara is the greatest source of mineral dust aerosol (see e.g. Koven & Fung, 2008 and Prospero, Ginoux, Torres, Nicholson, & Gill, 2002 for detailed discussions), there has been an increasing interest in the Gobi desert and other Central Asian dust sources in the recent decades, primarily in connection with air quality concerns and pollution in eastern Asia. Studies on the mineral dust in this region of the globe include, for instance, Chin et al. (2003) or Ma, Liu, Liu, Ma, and He (2012). Recent efforts to measure the refractive index of Saharan dust from several locations and under controlled conditions have been performed at the AIDA chamber of the Karlsruhe Institute of Technology by Wagner et al. (2012).

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Because of its significant impact of mineral dust aerosol in a variety of different areas, desert dust is a prominent component of existing databases for the optical properties of aerosols, such as OPAC by Hess, Koepke, and Schult (1998) or the database by Levoni, Cervoni, Guzzi, and Torricella (1997). Nevertheless, for mineral dust aerosol, the modeling of its optical properties has proven to be a formidable challenge, as the scattering dust particle is strongly heterogeneous and consists of many different mineral types and chemical species. Regional variability of the composition of mineral dust particles has been recognized in the past to further complicate the problem and studies on this topic are manifold, including the efforts by Formenti et al. (2011) and Di Bagio et al. (2014) and Ryder et al. (2013) and most notably by Dubovik et al. (2002) and Dubovik et al. (2006) with a focus on optical properties and radiative transfer.

Due to the random, heterogeneous morphologies of mineral dust particles, there is interdependence when it comes to the modeling of the shape and the refractive index of this particular type of aerosol in microphysical light scattering computations. It has been shown by Mishchenko, Travis, Kahn, and West (1997) that the nonsphericity of the dust scattering particles needs to be taken into account for single scattering calculations. Examples for the two different approaches which exist for determining the optical properties of mineral dust particles are the work by Kahnert (2004) on the one hand and the work by Liu, Panetta, Yang, Macke, and Baran (2013) and Ishimoto, Zaizen, Uchiyama, Masuda, and Mano (2010) on the other. While Kahnert uses simple geometrical particle shapes such as spheres and spheroids and considers the heterogeneity through the refractive index, Ishimoto develops a complex random morphology model with different refractive index values specified in several disjoint particle subdivisions based on a spatial Voronoi tessellation. The effective medium approach discussed in this manuscript follows the first philosophy and allows managing the complexity involved in considering both a regional and a size-dependence of the refractive index. Based on the optical properties, the quantification of the influence of dust on radiation transport has been the subject of several studies, including Myhre and Stordal (2001) and Sokolik et al. (2001). The general consensus is that the uncertainties in the dust refractive index are the dominant source of error in the aforementioned radiation transport calculations, as discussed for instance by Myhre and Stordal (2001). Consequently, the current model aims to improve upon this situation from multiple perspectives. The spectral dependency of the refractive index on the wavelength λ of the incident light is computed as a function of the particle size, and for the first time, a rough distinction between different mineral source regions is taken into account.

This paper is organized as follows: Section 2 is the refractive index model description, further expanded upon in Sections 3 and 4, which in particular discuss data acquisition and effective medium calculation respectively. Section 5 presents the analysis of the obtained spectra in terms of the Hilbert transform or the Kramers-Kronig integral, where the first name is rather exclusive to mathematics while the second is more often encountered in physics and material science. Section 6 discusses the computation of first and second moments based on AERONET aerosol size distributions, in order to compute mean refractive index spectra independent of the particle size.

2. Refractive index model description

The theoretical basis for the model developed in this study can be found in the work of Otto et al. (2009). To compute the solar radiative effects of a Saharan dust plume, these authors developed a refractive index model based on chemical composition data for northern Saharan dust as a function of particle geometrical size published by Kandler et al. (2009). The distinctive feature of the current model is a quite useful computational tool to take the size dependence and the regional variation as parameters influencing the refractive index into account by applying the same procedure as Otto et al. (2009) to different geographical regions where sufficient composition data of mineral dust is available.

Generally, the model requires the chemical composition of a mineral dust particle as an input. The composition needs to be provided in terms of the volume fraction of the particle's constituent minerals over the geometric diameter. Based on these volume abundance ratios, the refractive index spectra of the individual minerals can be combined in various ways. As argued by Kandler et al. (2009), the lack of cohesive refractive index spectra from different authors for many mineral components, as well as the overall sheer number of different mineral types further force the introduction of a simplified constituent model, where the individual mineral types are merged to overarching mineral classes such as e.g. a Silicate class combining Illite, Kaolinite and Chlorite. The different mineral classes into which the individual mineral types are classified are listed in Table 1 and are largely identical with the classification scheme proposed by Kandler et al. (2009). Additional substances included in the internal mixing, such as water, are considered as separate categories.

As an example, the relative volume abundance for the mineral groups in the northern Sahara as measured by Kandler et al. (2009) is shown in Fig. 1. It may be observed that the volume fraction of all components changes drastically versus particle diameter, except for the iron oxides, with low concentration remains without significant variations in magnitude.

The next necessary input for the model are the refractive index spectra of the individual mineral types. These spectra often are available only piecewise for certain bandwidths and/or refractive index measurements from different sources in the literature for the same mineral under identical conditions differ by a non-negligible amount. Consequently, a sufficiently large collection of spectra from different experimental investigations needs to be collected and combined to a global patchwork spectrum. A collection of different measurements from different authors is shown in Figs. 2 and 3 and the already mentioned issues regarding the refractive index data available in the literature may be observed directly. The figures show the real and imaginary refractive index spectra from the Silicate class, respectively. The sources for both the regional size-dependent composition data, as well as the measured refractive index spectra for the individual mineral components are given in the next Section.

As there is no robust criterion to determine the quality and plausibility of a specific measurement versus other counterparts, the data points have been interpreted as random noise and combined spectra have been filtered using a local regression method. In