Accounting for particle non-sphericity in modeling of mineral dust radiative properties in the thermal infrared

M. Legrand *, O. Dubovik, T. Lapyonok, Y. Derimian

Laboratoire d'Optique Atmosphérique, University of Lille 1, 59655 Villeneuve d'Ascq, France

ABSTRACT

Spectral radiative parameters (extinction optical depth, single scattering albedo, asymmetry factor) of spheroids of mineral dust composed of quartz and clays have been simulated at wavelengths between 7.0 and 10.2 μm using a T-matrix code. In spectral intervals with high values of complex index of refraction and for large particles, the parameters cannot be fully calculated with the code. Practically, the calculations are stopped at a truncation radius over which the particles contribution cannot thus be taken into account. To deal with this issue, we have developed and applied an accurate corrective technique of T-matrix Size Truncation Compensation (TSTC). For a mineral dust described by its AERONET standard aspect ratio (AR) distribution, the full error margin when applying the TSTC is within 0.3% (or $\pm 0.15\%$), whatever the radiative parameter and the wavelength considered, for quartz (the most difficult case). Large AR values limit also the possibilities of calculation with the code. The TSTC has been able to complete the calculations of the T-matrix code for a modified AERONET AR distribution with a maximum AR of 4.7 instead of 3 for the standard distribution. Comparison between the simulated properties of spheroids and of spheres of same volume confirms, in agreement with the literature, that significant differences are observed in the vicinity of the mineral resonant peaks (λ ca. 8.3–8.7 μm for quartz, ca. 9.3–9.5 μm for clays) and that they are due to absorption by the small particles. This is a favorable circumstance for the TSTC, which is concerned with the contribution of the largest particles. This technique of numerical calculation improves the accuracy of the simulated radiative parameters of mineral dust, which must lead to a progress in view of applications such as remote sensing or determination of energy balance of dust in the thermal infrared (TIR), incompletely investigated so far.

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1. Introduction

A mineral dust layer in the atmosphere constitutes a barrier limiting (i) the downward propagation of solar radiation to Earth surface and (ii) the upward propagation of terrestrial radiation, emitted primarily by Earth surface, to space. Through this double direct radiative effect, mineral dust appears as a significant potential contributor to the change of the energy balance of the planet. It scatters a fraction of solar radiation to space (parasol effect) and a fraction of terrestrial radiation emitted by the surface – leading in addition to a heating of the surface and of the lower atmosphere (reinforcing the greenhouse effect from gases, exactly like cloud cover). Thus it is of the

* Corresponding author. Tel.: +33 320436646.
E-mail addresses: michel.legrand@univ-lille1.fr (M. Legrand),
oleg.dubovik@univ-lille1.fr (O. Dubovik),
tatsiana.lapionak@univ-lille1.fr (T. Lapyonok),
yevgeny.derimian@univ-lille1.fr (Y. Derimian).

highest interest to study the optical properties of mineral dust, not only for its impact on solar radiation as regularly emphasized in the scientific literature, but also on terrestrial radiation in the thermal infrared (TIR) spectrum (wavelengths from 3 to 20 μm), especially in the 10-μm atmospheric window where radiative transfer from the planet to space can take place efficiently.

Mineral dust is (with sea salt aerosol), one of the most abundant and widespread components of the global aerosol system. Its global production ranges between 1000 and 3000 Tg yr^{-1} [1]. The present study is focused on the analysis of the desert dust – a subcategory of mineral dust representing most of it (however, in this paper both terms are interchangeable in most cases; frequently, using "dust" is sufficient). Desert dust is a mixture of particles of variable complexity, having different sizes and shapes, constituted of various minerals, generally crystalline, originated in erodible desert soils.

These soils are classified according to their size texture as clay (grains smaller than 2 μm according to the USDA classification), silt (grains between 2 μm and 50 μm) and sand (grains larger than 50 μm). These three basic soil textures are then used to describe the actual soils as mixtures between them in variable fractions.

The erodible desert soils are also characterized by their mineral composition in relation with the local geological features at the surface and the origin of the sediments transported by wind and water. A consequence is that desert dust composition varies with geographical location of the source where it is originating (see [2 and 3] on the mineralogical signature of the African sources). Different minerals do not appear in the same size ranges of the dust particles, corresponding to the fact that clay, silt and sand grains have not the same mineral composition [4]. The sources composed of these erodible soils are activated through wind deflation by processes of grains creeping and saltation, aggregates sandblasting and final dust particles emission [5], leading to granulometric fractionation. As the soil size texture depends on the mineral composition, a mineralogical fractionation occurs in association to the granulometric fractionation through the processes resulting in dust emission.

The surface wind speed makes an impact not only on the dust amount uplifted in the atmosphere, but also on the particle size distribution and composition. It has been shown theoretically that a stronger surface wind provides a larger kinetic energy to the grains in saltation leading to emission of smaller particles through the sandblasting process and increasing the clay fraction [6]. This has been verified in field experiments, confirming and explaining the presence of a sub-micron mode for dust (between 0.5 and 0.1 μm) [7]. On the other hand, it has been proposed by Thomas and Gautier [8], that the larger quartz fraction observed in dust storms when compared with background dust in a same region (using a compilation of the literature, especially from [9]) would be due to a higher efficiency of the strong dust storm winds to mobilize the heavier and more massive quartz particles in the surface soil (compared with the clay particles). These reported effects of the surface wind speed are apparently contradictory, but they could be explained by the fact that the large quartz amount lifted by strong winds of a dust storm

is observed only for fresh dust insofar as the wind decay leads to a quick settling of the large quartz particles.

Further evolutions of the size distribution and of the mineral composition during dust transport arise from (i) gravity settling [10], (ii) chemical reactions with atmospheric components, (iii) coalescence with other aerosol species, (iv) the interactions with water (vapor, liquid, ice).

The ensemble of mineral species present in a dust sample (see [11] for a description), determine the radiative properties of this dust as a combination of the radiative properties of each constituent mineral. The phylosilicates (kaolinite, illite, montmorillonite, chlorite, palygorskite) proceed from clay soil. The main other minerals are silica (quartz crystal or amorphous), calcium carbonate (calcite crystal or amorphous), calcium sulfate (gypsum crystal or amorphous), iron oxides (hematite, goethite), aluminum silicates (crystals of alkali and plagioclase feldspars), proceeding principally from silt soil and to a lesser extent from clay and fine sand.

The radiative properties of dust are to a large extent defined through the complex refractive index spectra of the constitutive mineral species. In comparison with the solar spectra, in the TIR these spectra depend strongly on the minerals of concern and they are usually highly variable with the wavelength for each mineral. This is the case as well for the resulting radiative properties of these minerals (compare between solar and TIR refractive indices, or extinction coefficients, in [12]). Particularly, extinction and absorption spectra of these minerals show resonances as peaks, frequently located at wavelengths in the 10-μm atmospheric window, which can be used for the identification of each mineral.

The analyses of dust composition in the laboratory are carried out on dust samples deposited on filters. The wavelength dispersive X-ray fluorescence and the particle-induced X-ray emission are techniques directly applicable to the dust samples, leading to the quantitative elemental composition of each sample [13]. But the relation between elemental and mineral compositions is usually complicated, especially for dust samples containing many minerals, making the mineral analysis approximate and possibly incomplete. However, such elemental analysis can be applied to individual particles by means of electron microscopy [14–17]. Such approach is then helpful to identify and measure the amount of certain minerals. For example, a Ca/S ratio close to 1 for a high S content indicates the presence of calcium sulfate in the particle, involving primarily gypsum presence and subsidiarily amorphous sulfate arising from the atmospheric reaction of calcium carbonate with sulfur dioxide or sulfuric acid (the method cannot discriminate between crystalline and amorphous states of the mineral). Another example is the use of the Si/Al ratio to determine whether a phylosilicate particle would chiefly contain illite (ratio greater than 2) or kaolinite (ratio close to 1) [17]. The limitation of the particle numbers processed by use of the electron microscopy (order of 10^2 to 10^3) has been recently resolved by automated recognition procedures enabling to consider much larger numbers (order of 10^4 to 10^5) [18,19].

Another approach is the direct identification and quantitative analysis of the crystalline minerals constituting the