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SEPARATION OF FINE AND COARSE AEROSOL CONTRIBUTIONS TO THE TOTAL AEROSOL LIGHT SCATTERING: AN AERONET ASSESSMENT

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ABSTRACT. The contribution of each mode to total light scattering in a bimodal aerosol population is studied. The dependence of some backscattering-related light properties on particle shape, and concentration, are simulated. Results will help us determine whether any of those parameters will be useful in separating the optical properties of each particle mode, and therefore in determining mode concentration.

1. Introduction

Mineral particles play an important role in the Earth's radiation budget. The long-range transport of mineral particles by the combined action of convection currents and general circulation systems make these particles a significant constituent even at locations far from their sources [1]. However, studies of natural aerosols are few compared to those of anthropogenic aerosols. In last decade, several extensive investigations and coordinated field campaigns have been carried out to assess the impact of dust particles on radiative forcing. In Southern Europe, dust episodes are commonplace due to its proximity to the Sahara desert, one of the world's largest dust sources. Other parts of Europe felt the dire need to account for dust outbursts in 2010, when the Icelandic Eyjafjallajkull volcano forced the closure of the European airspace. Large dust sources can also be found in India (Thar desert), China (Taklamakan desert), the African Sahel (Bodele depression), etc. In all these cases, dust aerosols are not found alone, but usually form a mixture with other particles produced in combustion process (forest fires, fossil fuel combustion, biomass burning, urban traffic) [2]. The resulting particle size distribution (PSD) could be described as a mixture of a submicron-sized fine mode of combustion-produced particles, and a micron-sized coarse mode of dust particles.

Photometric measurements of solar extinction and sky radiance are used in inversion models to yield information on columnar-integrated aerosol particle size distribution, particle concentration, refractive index, single scattering albedo or asymmetry parameter [3, 4]. Such inversions do not yield any information on particle shape, or on the refractive index of either mode (an average value is given). Neither it includes other light-scattering parameters such as backscattering, depolarization ratio, or lidar ratio; parameters of great

Table 1. Particle size distribution parameters.

	Mode 1 (fine)	Mode 2 (coarse)
Concentration $C_{v,i}$ ($\mu m^3/\mu m^2$)	0.080	0.078
Modal radius $r_{mod,i}$(μm)	0.205	1.721
Width σ_i	0.485	0.631

importance in particle characterization as they are highly dependent on particle shape. On the other hand, multi-wavelengths lidars allow an independent calculation of aerosol extinction and backscattering coefficients profiles, together with particle depolarization ratio. Microphysical properties of particles have been obtained from remote sensing techniques by assuming spherical particles, with some degree of success [5]. But the effect of particle shape cannot be discarded in some parameter like backscattered depolarization ratio, which has a zero value for spherical scatterers. Last year Veselovskii et al [6] adopted the AERONET inversion concept for the incorporation of a spheroidal model into the lidar retrieval of aerosol physical properties.

In the present paper, an attempt is made to separate fine and coarse mode contributions to the total aerosol light scattering. AERONET data (PSD, refractive index, concentration) have been used as input, and computer simulations have been carried out to find out what light scattering parameters are most sensitive to changes in particle shape, as well as in relative fine/coarse concentration. Results are column-integrated, but future applications will include a layer-by-layer separation profiling.

2. Method

Particle size and composition have been chosen to match the AERONET direct measurements and inversion data for the Eyjafjallajkull outburst as measured on April 19, 2010, 14:49:37 GMT (ITF-Leipzig Station). Values of the refractive index used for simulations are m=1.459+i0.0036 (λ=532 nm), and m=1.478 + i0.0022 (λ=1064 nm). The particle distribution has been modeled as a sum of two lognormal volume size distributions:

$$\frac{dV(r)}{d\ln r} = \sum_{i=1}^{2} \frac{C_{v,i}}{2\pi\sigma_i} \exp\left[-\frac{\ln(r/r_{mod,i})^2}{2\sigma_i^2}\right] \tag{1}$$

where $C_v = C_{v,1} + C_{v,2}$ is the total volume concentration per unit lateral area. Radius and width (r_{mod},s) values have been selected according to the AERONET-inverted data: (0.205 μm, 0.485) for the fine mode, (1.721 m, 0.631) for the coarse mode, as given in Table 1. As particles are assumed to be non-spherical, any radius parameter refers to the equivalent-volume-sphere radius. Particles for both modes (fine and coarse) have been assumed to be a 50-50 volume mixture of prolate and oblate spheroids with the same axial (long-to-short axis) ratio e. For each mode, varying e values have been considered, ranging from 1.2 to 1.8. The former value is a good approximation to the average eccentricity of dust aerosols [7], while the latter covers the case of close-to-sphericity particles. An axial ratio of 1.0 was not deemed adequate because perfect sphericity is hard to find in aerosols, and even a small amount of nonsphericity can make a large contribution to depolarization and lidar ratio values.

Computer simulations have been carried out by using ALFA, a pre-calculated database of light-scattering data for non-spherical particles, based on the T-matrix method [4] and the Dubovik approach for large-scale database computations [8]. Two wavelength values, 532nm and 1064 nm, have been used.

3. Results

Calculations for several values of the axial ratio in both fine and coarse modes showed a 75% variation in the backscattering coefficient at 532 nm (65% for 1064 nm). The backscattering coefficient ratio t(532)/t(1064), or color ratio, has a smaller variation (29%), with the highest value for a quasispherical fine mode and a highly nonspherical coarse mode. Lidar ratio (extinction to backscattering ratio) also changed within a 70% range from the smaller to the largest value, with increasing values as both fine and coarse mode particles departed further from sphericity. It has been found that lidar ratio values for λ=1064 nm do not depend on the axial ratio of the fine mode, and could therefore be used to determine the shape of the particles in the large (coarse) mode. Depolarization ratio values also had a large dependence on shape, with differences of up to 160% for 532 nm wavelength, and 57% for 1064 nm.

In order to determine whether backscattered light-scattering parameters can be useful as a tool to separate fine and coarse modes, the relative concentration of the fine mode (C_{rf}) was changed from 0 (only coarse mode) to 1 (only fine mode), while keeping the total volume concentration fixed to 0.08 m^3/m^2. It was found that the color ratio curve rises monotoonically for increasing concentration of the fine mode. The change becomes linear for a coarse mode axial ratio e_2=1.6.

Depolarization ratio values at 1032 nm show little dependence on fine mode particle shape e_1, and for higher fine mode concentrations (C_{rf}<0.3) a weak dependence on coarse mode particle shape e_2 is also found. Lidar ratio values behave differently, but show no dependence on $e_{1.}$ On the other hand, data for 532 nm show a different behavior, with a clear influence of fine mode concentration and axial ratio values for both modes. Depolarization and lidar ratio values at both wavelengths can therefore then be used to obtain a reasonable value of relative mode concentrations, if particle size and composition are assumed.

4. Concluding remarks

Light-scattering parameters depend on particle size, shape, and composition in a very complex way. Some quantities show little dependence on shape, (e.g. extinction optical depth, single-scattering albedo), while those related to backscattering light, are more sensitive to both shape and relative concentration. Results in this paper cannot be extrapolated automatically to other size distributions of aerosols, but give us a hint of which parameter can be best put to use in order to determine either shape or relative concentration in a two-modal population of soot/dust particles. This is particularly useful in lidar measurements, where experimental data can be obtained for a set of horizontal layers, as well as for the entire column. Relative concentration of both modes could be determined from backscattered parameters, and thus a separation of fine and coarse mode could be made. This approach is similar to that of [9], with the difference that the input data will not be taken from the literature, but calculated in a rigorous way via the T-matrix method.

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