ALFA: A Database for Light Scattering Simulations with Atmospheric Aerosol Applications

ALFA: Una Base de Datos para Simulaciones de Dispersión de Luz con Aplicaciones en Aerosoles Atmosféricos

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ABSTRACT:

An extensive database of light scattering (LS) properties for nonspherical particles has been computed to help in the radiative-transfer calculations on atmospheric aerosols. Its main features and size/shape/composition range are described. A second database, with kernel functions, has also been calculated to simplify the modeling of polydisperse particle size distributions.

Key words: Light Scattering, Aerosol, T-matrix, Database, Nonsphericity

RESUMEN:

Se ha confeccionado una extensa base de datos sobre propiedades de dispersión de luz por partículas esféricas, con el objetivo de servir de ayuda para el cálculo de las propiedades de transferencia radiativa en atmósferas con aerosoles. Se describen sus principales rasgos y el rango de valores de tamaños, forma y composición. Ello permitió calcular una base de datos con funciones kernel, que permite simplificar el modelado para distribuciones de tamaño de partículas.

Palabras clave: Dispersión de Luz, Aerosol, Matriz-T, Base de Datos, No-esfericidad

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1. Introduction

The use of the radiative-transfer equation (RTE) to model light-scattering (LS) parameters of atmospheric aerosol populations requires the calculation of single-scattering quantities such as the extinction and scattering cross sections (C_{ext}, C_{sca}), the asymmetry parameter, and the Müller matrix $\mathbf{P}(\theta)$ that relates the Stokes vectors of incident and scattered radiation. Mie theory for spherical scatterers is often used with success, but in atmospheric aerosols particle shape typically takes a non-spherical shape, which calls for the use of non-spherical LS methods.

In order to model nonspherical aerosols, particles will be represented as a collection of scatterers with a symmetry plane (e.g. spheroids). In that case, only six Müller matrix elements are nonzero, which can be expanded into a set of generalized spherical functions [1,2]. The expansion coefficients of such expansion depends on the particle parameters but not on the incident of scattered light, so they can be used to build the Mueller matrix for any scattering direction.

Light scattering properties of a polydisperse collection of scatterers can be modeled as size averages of monodisperse data. Optical coefficients and Mueller matrix elements can be calculated as [3]:

$$\begin{aligned} \tau_{ext,sca}(x,m,r,e) &= \int_{r_{\min}}^{r_{\max}} C_{ext,sca}(m,x,e)\frac{dN(r)}{dr}dr \\ \tau_{sca}P_{ij}(\lambda,m,r,e,\theta) &= \int_{r_{\min}}^{r_{\max}} C_{ij}(m,x,e,\theta)\frac{dN(r)}{dr}dr \end{aligned} \qquad (1)$$

where $m=n+ik$ is the (complex) index of refraction; r,e are size and shape parameters; $x=kr$ is the dimensionless size parameter for a wavelength $\lambda=2\pi/k$; and N(r) is the number size distribution of particles.

2. Method

2.a. The need for kernel functions

In order to obtain optical depth and other light scattering parameters, a set of cross sections and Mueller matrix elements are to be calculated for a wide range of size, shape, and composition values. For a given atmospheric aerosol event, this can quickly become a cumbersome and time-consuming task. In order to alleviate this problem, sets of so-called kernel functions can be used. It is based on the fact that equation (1) can be written as:

$$\tau_{sca} \approx \frac{2\pi}{\lambda}\sum_{p=1}^{N_p}\sum_{l=1}^{N_l}\frac{dV(r_p)}{dlnr}\frac{dN'(e_l)}{dlne}K_{sca}(m,x_p,e_l) \qquad (2)$$

[4], where dV/dlnr denotes volume size distribution of particles. The so-called kernel functions K_{sca} depend on the index of refraction m, and the size/shape parameters x_p, e_l. Kernel functions for scattering, asymmetry, backscattering, and Mueller matrix elements can be calculated in a similar way. This approach requires a precalculated set of kernel functions for (N_p, N_l) size and shape values, for a given value of the index of refraction m.

The number of size bins (N_p) will determine the usefulness of the approximation given by equation (2). Low N_p values means a small number of bins and increasing errors. On the other hand, an excessively large number of very narrow bins will increase the number of kernel functions without an appreciable gain in accuracy. The number of integration points in equation (2) will also influence the accuracy of kernel function computations.

2.b.- ALFA/BETA

To calculate a set of kernel functions for atmospheric aerosol modeling, single scattering data must be computed for a number of size, shape, and index of refraction values. In the present work, scatterers are modelled as spheroids (both prolate and oblate) with an equal-volume-sphere radius r_{eq}. The particle size and shape will be characterized by two parameters: the axial ratio e and the equivolume (dimensionless) size parameter $x_{eq}=k*r_{eq}$, where $k=2\pi/\lambda$ is the wavenumber for a wavelength λ. Light scattering properties have been calculated using the T-matrix method [2].

The monodisperse size parameter values were logaritmically spaced as $\ln x_i = \ln x_o + i\Delta\ln x$ (i=1 to 4100). This calls for the calculation of 41 bins (x=0.01, 0.012589 … 100), each with a width $\Delta\ln x$ =0.012589. The maximum size parameter used in the calculation process (x=125.89) yields an equivolume-sphere radius of about 8 microns at a wavelength of 500 nm. This is not easy to achieve, particularly for highly elongated particles, where the T-matrix fails due to convergence problems. For oblate spheroids with an axial ratio 3, for instance, the maximum size value falls to about 25-30 (r_{eq} about 2-2.5 microns).

The shape range has been set by assuming a total of 10 axial ratio values for each form (oblate and prolate): e=1.2, 1.4, … 3.0. Regarding composition, a total of 225 values of the index of refraction $m=n+ik$ were chosen, ranging from 1.33 to 1.6 for the real part, and 0.0005 to 0.5 for the imaginary part. Both ranges were also logaritmically spaced:

n=1.33, 1.3477 … 1.579, 1.6
k=5*10^{-4}, 8.189*10^{-4} … 0.3050, 0.5

This adds up to a maximum total of 4101*20*225 = 18.454.500 different size/shape/composition values. A large set of calculations was made which includes cross sections and expansion coefficients a_i^s, b_i^s (eq. 3) for all those particles. The result is the ALFA database.

The full computation of ALFA in its current form took about one full year of computation in several kinds of machines, from PCs to supercomputers available through the Spanish Supercomputation Network (RES, Red Española de Supercomputación). As stated stated above, calculations in the entire size range x=0.01 to 125.89 was not feasible at all values of the shape parameter, which means ALFA is incomplete at this stage.

Even in the current, incomplete, state, ALFA is now operational. It has allowed to compute a set of kernel functions (eqs. 2), making up a second database called BETA. The validity of the approximation in equation 2 (and, therefore, the value of BETA as a calculation tool) has been checked with data from the OPAC database for spherical particles [5]. Our calculations by direct integration from ALFA data agree with the OPAC data to the last digit. Results calculated by using the BETA database (equation 2), fit the OPAC data to within 0.5%.

2.c.- Future developments

Tests with experimentally-obtained data are under way to evaluate the best strategy to use our pre-calculated databases. Several conclusions are being drawn from day-to-day usage experience. For example, some simulation procedures assume equal number of oblate and prolate scatterers with the same axial ratio. Therefore, it became convenient to enlarge both ALFA and BETA in order to cover LS data for a 50-50 (in volume) mixture of oblate and prolate particles. This was done with relative ease from existing data. CEAMA equipment now in place can measure quantities like depolarization ratio and Angstrom coefficient, which also called for the confection of tailor-made subset databases. Additionally, some authors use number (not volume) particle size distributions, so new kernel functions had to be calculated.

Other database additions include spherical shape for comparison, as well as additional values for the index of refraction. The need to obtain subsets for small sets of refractive index, shape, and/or particle size distribution values has also arisen, and will require special computer codes for data manipulation. A recently-developed set of computer codes also allow for the direct manipulation of ALFA monodisperse data in order to do calculations on monomodal and bimodal particle size distributions. Bimodal PSDs are particularly important in atmospheric aerosols where two modes (fine and coarse) can be assumed.

As an example of the ALFA usage in light scattering calculations, depolarization ratio values for randomly-oriented oblate and prolate spheroids with several axial ratios are shown in Figure 1. The index of refraction assumed used (m=1.4978+i0.009653) is related to dust particles. Particle size values follow a lognormal volume particle size distribution function, where r is the radius of the equivalent-volume sphere, is for a modal radius r_{mod} and width σ:

$$\frac{dV(r)}{d\ln r}=\frac{C_v}{\sqrt{2\pi}\sigma}\exp\left[-\frac{\ln(r/r_{\text{mod}})^2}{2\sigma^2}\right] \qquad (4)$$

In order to make Figure 1 wavelength-independent, the dimensionless size parameter $x_{\text{mod}}=(2\pi/\lambda)r_{\text{mod}}$ has been used in the X axis; Y axis shows the depolarization ratio, defined as the ratio of the flux of the cross-polarized component of the backscattered light relative to that of the co-polarized compoment.

Figure 1 shows how depolarization ratio values shows a heavy dependence not only on axial ratio (as it was expected, backscattering being shape-dependent), but also on the shape of the particle, showing that an equal-volume mixture of oblate and prolate spheroids with the same axial ratio is not always a valid assumption. Similar data for m=1.4978+i0.5 (not shows here) yields a depolarization ratio which, while small, depends strongly on the particle shape and axial ratio. The fact that highly-absorbing material is a weak depolarizer can be of importance un atmospheric aerosols composed of two particle populations like soot and dust.

ALFA also allows for practical applications, as Figure 2 show. Extinction coefficient values have been simulated for the 2009 Eyjafjallajökull volcano dust plume. Particle size distribution values were measured on May 6, 2009 (11:00 h UTC) by a plane near León (north Spain), and have been used to calculate extinction at three wavelengths: 355, 532, and 1064 nm. Number concentration has been calculated as 91.9 particles/cm^3 Two different values of the index of refraction were assumed: 1.534+i0.0035 and 1.558+i0.0059; particles were modelled as a 50% mix of oblate and prolate spheroids with equal axial ratio.

3. Conclusions

A database has been built and tested to alleviate the burden of computer simulations on atmospheric aerosols, by calculating light scattering data for a wide range of particles of interest. This is now possible due to the increasing power of modern computer equipment, as well as a greatly improved storage capacity (ALFA storage requirements are about 45 GBytes in its current stage). Parameters obtained by ALFA include optical cross sections, phase function and polarization ratios, and can be directly applied in aerosol modeling. A second, derived database containing kernel functions (BETA) is useful in radiative-transfer-equation solving.

Future developments are under way to include light scattering calculations to cover size/shape values currently unavailable to the standard T-matrix method. Additional options under study include geometrical optics approximation and the development of a T-matrix scheme using spheroidal coordinates. Even in its incomplete form, however, ALFA is being used for light-scattering simulations on particle size distributions of atmospheric aerosols.

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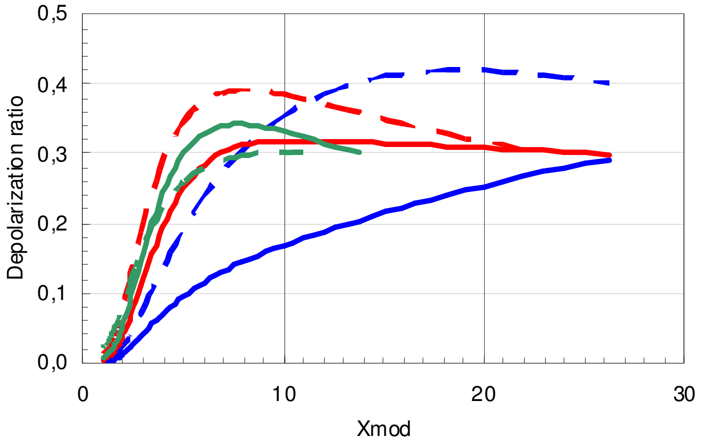

Fig. 1. Depolarization ratio values for several populations of particles, each with a different shape (full line: oblate spheroids; dashed line: prolate spheroids) with axial values ε:1.2 (blue), 1.6 (red), and 2.0 (green). A volume log-normal particle size distribution (witdh σ=0.5), and an index of refraction value *m*=1.4978+i0.009653, are assumed.

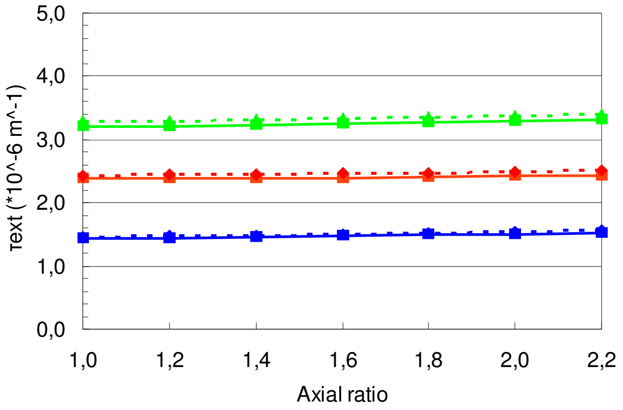

Fig. 2. Simulation of extinction coefficiente values, assuming particle size distribution equals to that measured in León (northern Spain) at 11:00h UTC, May 6, 2009 Eyjafjallajökull volcano dust plume. Two values of the index of refraction (full line: 1.534+i0.0035; dashed line:1.558+i0.0059) are assumed for wavelengths 355 (green), 532 (red), and 1064 (blue) nanometers.