ALFA/BETA: A Dual Database for Light Scattering Simulations on Atmospheric Aerosols

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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/range/composition range are described. A set of kernel functions database has also been calculated to simplify the modelling of polydisperse particle size distributions.**

Keywords— **Light Scattering, Aerosol, Database, T-matrix, nonsphericity.**

I. 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, but in atmospheric studies particle shape typically takes a non-spherical shape, which calls for the use of non-spherical LS theories.

In order to model nonspherical aerosols, the assumption will be made that such particles can be represented as a collection of scatterers with a symmetry plane (e.g. spheroids). In such case, the Müller matrix takes the form [Bohren and Huffman, 1983]:

$$P(\theta)=\begin{bmatrix} P_{11}(\theta) & P_{12}(\theta) & 0 & 0 \\ P_{12}(\theta) & P_{22}(\theta) & 0 & 0 \\ 0 & 0 & P_{33}(\theta) & P_{34}(\theta) \\ 0 & 0 & -P_{34}(\theta) & P_{44}(\theta) \end{bmatrix} \quad (1)$$

where the matrix elements are typically normalized as:

$$\frac{1}{2}\int_0^{\pi} P_{11}(\theta)\sin\theta d\theta = 1 \quad (2)$$

Each matrix element can be, in turn, expanded into a set of generalized spherical functions [Mishchenko, 1991]:

$$\begin{aligned} P_{11}(\theta) &= \sum_{s=0}^{\infty} a_1^s P_{00}(\cos\theta) \\ P_{22}(\theta)+P_{33}(\theta) &= \sum_{s=2}^{\infty} (a_2^s + a_3^s) P_{22}(\cos\theta) \\ P_{22}(\theta)-P_{33}(\theta) &= \sum_{s=2}^{\infty} (a_2^s - a_3^s) P_{2-2}(\cos\theta) \\ P_{44}(\theta) &= \sum_{s=0}^{\infty} a_4^s P_{00}(\cos\theta) \\ P_{12}(\theta) &= \sum_{s=2}^{\infty} b_1^s P_{02}(\cos\theta) \\ P_{34}(\theta) &= \sum_{s=2}^{\infty} b_2^s P_{02}(\cos\theta) \end{aligned} \quad (3)$$

The expansion coefficiente a_i^s, b_i^s, have the advantage of being dependent 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. The extinction and scattering optical depths can be calculated as [Dubovik and King, 2000]:

$$\begin{aligned} \tau_{ext,sca}(x,m,r,e) &= \int_{r_{\min}}^{r_{\max}} C_{ext,sca}(m,x,e)\frac{dN(r)}{dr}dr \\ \tau_{sca}P_{ij}(\lambda,m,r,e,\theta) &= \int_{r_{\min}}^{r_{\max}} C_{ij}(m,x,e,\theta)\frac{dN(r)}{dr}dr \end{aligned} \quad (4)$$

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. Likewise for the Mueller matrix elements. An additional shape distribution N'(e) can be added in order to account for different particle shapes.

II. METHOD

A. The need for kernel functions

In order to obtain optical depth and other light scattering parameters (eq 4), 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.

LS properties by an ensemble or particles typically depend on the particle volume (or surface) rather than the number concentration, and cross sections usually show a smoother variability for equal relative steps $\Delta r/r$ that for equal absolute steps Δr. Therefore, it is more convenient to rewrite equation (4) as:

$$\tau_{sca}(\lambda,m,r,e)=\int_{Ln\,r_{\min}}^{Ln\,r_{\max}}\frac{C_{sca}(m,x,e)}{v(r)}\frac{dV(r)}{dlnr}dlnr \qquad (5)$$

[Dubovic et al, 2006], where v(r) is the volume of the particle with size parameter r, and dV/dlnr denotes volume size distribution of particles. It is equivalent to eq (4), but has fewer integration points and is easier to calculate.

Next, we approximate eq. (5) as:

$$\tau_{sca}\approx\frac{2\pi}{\lambda}\sum_{p=1}^{N_p}\sum_{l=1}^{N_l}\frac{dV(r_p)}{dlnr}\frac{dN'(e_l)}{dlne}K_{sca}(m,x_p,e_l) \qquad (6)$$

where the 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. As an example, the phase function can be approximated as

$$\tau_{sca}P_{11}\approx\frac{2\pi}{\lambda}\sum_{p=1}^{N_p}\sum_{l=1}^{N_l}\frac{dV(r_p)}{dlnr}\frac{dN'(e_l)}{dlne}K_{11}(m,x_p,e_l,\theta) \qquad (7)$$

This approach requires a precalculated set of kernel functions for (N_p, N_l) size and shape values, for a given value of m. Kernel functions can be obtained by integration in a narrow size and shape interval called *bin*:

$$K_{sca}(m,x_p,e_l)=\int_{Ln\,e_l}^{Ln\,e_{l+1}}dlne\int_{Ln\,x_p}^{Ln\,x_{p+1}}\frac{Q_{sca}(m,x,e)}{v(x)}dlnx \;;\; \int_{Ln\,x_p}^{Ln\,x_{p+1}}\frac{Q_{sca}(m,x,e_l)}{v(x)}dlnx \qquad (8)$$

The number of size bins (N_p) will determine the usefulness of the approximation given by equation (8). Low N_p values means a small number of bins and a failure of the approximation in equation (8). 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 (8) will also influence the accuracy of kernel functions.

B. ALFA/BETA

In order to calculate a set of kernel functions to adequately model atmospheric aerosols, single scattering data must be calculated for a number of size, shape, and index of refraction values. In the present work, it is assumed that scattering particles are spheroids (both prolate and oblate) with long and short axes a,b, an axial ratio $a/b=e$, and 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 by using the T-matrix method [Mishchenko, 1991].

The monodisperse size parameter values were logaritmically spaced as $\ln x_i = \ln x_o + i\Delta\ln x$ (i=1 to 4100, $x=x_{eq}$). This calls for the calculation of 41 bins (x=0.01, 0.012589 … 100), each with a width set as $\Delta\ln x$ =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 calls for a maximum total of 4101*20*225 = 18.454.500 different size/shape/composition. A large set of calculations was made which includes cross sections and expansion coefficients (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 all kind of machines, from PCs to supercomputers available through the Spanish Supercomputation Network (RES, Red Española de Supercomputación). As 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.

Even in the current, incomplete, state, ALFA is now operational. It has allowed to compute a set of kernel functions (eqs. 8), making up a second database called BETA. The validity of the approximation in equation 8 (and, therefore, the value of BETA as a calculation tool) has been checked with data from the OPAC database for spherical particles [Hess *et al.*, 1998]. Our calculations by direct integration from ALFA data (equation 4) agree with the OPAC data to the last digit. Results calculated by using the BETA database (equation 8), fit the OPAC data to within 0.5%.

III. Present and 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.

Other database additions, now under calculation, include spherical shape, 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 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 to assume in atmospheric aerosols, where two modes (fine and coarse) are typically assumed.

As an example, figure 1 shows the relationship between asymmetry parameter and backscattering fraction (the fraction of light scattered in the 90º-180º region), of great importance to determine the size and composition of atmospheric aerosols. In this figure, the effect of axial ratio is shown, allowing us to assess whether the degree of nonsphericity is a relevant factor.

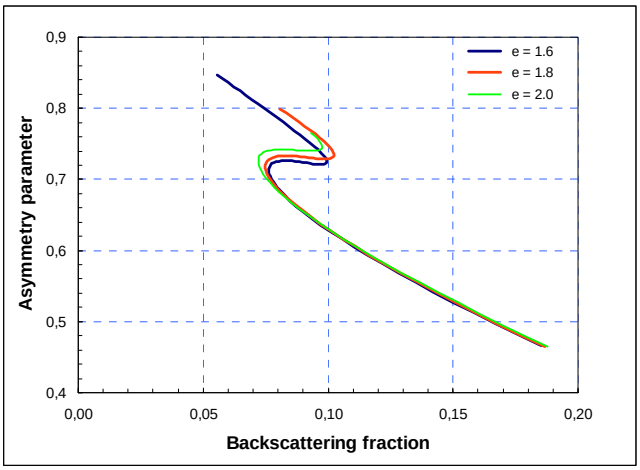

Fig. 1 Asymmetry parameter vs. backscattering values for a mix of oblate and prolate particles with axial ratios 1.6, 1.8, and 2.0, and an index of refraction m=1.498+i0.0097. Log-normal volume size distribution was assumed (σ=0.5, varying modal size parameters).

Future developments are under way on the ALFA/BETA database duo to include the extension of calculations to cover size/shape values currently unavailable to the standard T-matrix method.

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