APPLICATION OF RANDOMLY ORIENTED SPHEROIDS FOR RETRIEVAL OF DUST PARTICLE PARAMETERS FROM MULTI-WAVELENGTH LIDAR MEASUREMENTS

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ABSTRACT

Desert dust is responsible for a significant part of the total atmospheric aerosol loading, thus development of methods for the remote study of dust particle microphysical properties is demanded. Multi-wavelength (MW) Raman lidars have demonstrated their potential to profile particle parameters, however up to the present, the physical models used in retrieval algorithms for processing MW lidar data have been based on Mie theory. This approach is applicable for modeling light scattering by spherical particles only and does not adequately reproduce the scattering by generally non-spherical desert dust particles. Here we present an algorithm using a model of randomly oriented spheroids for inversion of multi-wavelength lidar data. Following the positive experience of retrieval developments within AERONET, we model aerosols as a mixture of two aerosol components: one composed only by spherical and second composed by non-spherical particles. The algorithm was tested with experimental data from a Saharan dust outbreak episode, measured with the BASIL multi-wavelength Raman lidar in August 2007.

1. INTRODUCTION

Desert dust aerosols play an important part in the Earth's radiation budget. A significant amount of information about the height distribution of dust particles has been gained during the last decade by means of Raman lidars (see [1] and references therein). The simultaneous detection of elastic and nitrogen Raman backscatter from lidar allows independent calculation of particle extinction and backscattering coefficients. Capabilities of Raman lidars are extended when the measurements are performed at multiple wavelengths. With this information, microphysical properties of aerosols can be retrieved through mathematical inversion [2,3]. However an application of these multiwavelength algorithms to dust measurements must overcome certain obstacles. For example, all existing lidar algorithms model the aerosol as an ensemble of spherical particles even though it is well established that backscattering by the particles of irregular shape is lower than predicted by Mie theory for spheres of equivalent volume. Moreover, the lidar backscatter from dust particles is strongly depolarized. The spectral dependence of particle depolarization ratio contains important information about particle parameters, but in the framework of Mie theory this information can't be exploited in the retrieval. Therefore the importance of accounting for particle non-sphericity in lidar retrievals today is well understood, however until now this effect remains under accounted in all known lidar algorithms.

Here we present the algorithm using a spheroid model for inversion of multi-wavelength lidar data. The model was suggested by Mishchenko et al., showing that the set of randomly oriented spheroids can adequately reproduce the phase functions measured for desert dust [4]. Dubovik et al. has developed an approach allowing fast and accurate computations of spheroid ensemble scattering based on pre-calculated look-up-table of scatterin kernels [5]. The model has been included in the AERONET retrieval algorithm in order to account for the effects of aerosol particle shape irregularities. More than five years experience with this AERONET code demonstrated the essential improvements in retrieval of dust properties. Here we adopt the same concept for including spheroid model into lidar retrieval. The performance of the algorithm is tested with experimental data from a Saharan dust outbreak episode, measured with the BASIL multi-wavelength Raman lidar in August 2007.

2. ALGORITHM DESCRIPTION

The main features of our previously developed algorithm [3] are used here. The optical data are related to the particle volume distribution by the Fredholm integral equations of the first kind:

$$g_i(\lambda_k) = \int_{\ln r_{min}}^{\ln r_{max}} \frac{C_i(m,r,\lambda_k)}{v(r)} \frac{\partial V(r)}{\partial \ln r} d\ln r \qquad (1)$$

$i = \alpha$, β_P, $\beta_\perp$; $k = 1,\dots, n$. Here the $g_i(\lambda_k)$ describe the optical data at the measurement wavelengths λ_k. The subscript i denotes particle extinction (α) and particle co- (β_P) or cross-polarized ($\beta_\perp$) backscatter coefficients; r is radius of the particle (for spheroids it is the radius of the sphere with equivalent volume). Radii r_{min} and r_{max} determine the lower and upper integration limit, $C_i(m,r,\lambda_k)$ denote the cross sections of extinction and backscattering and m is the complex refractive index. Using index p, which summarizes the

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kind of optical data (i) and wavelengths λ_k, Eq. (1) can be rewritten as:

$$g_p = \int_{\ln r_{\min}}^{\ln r_{\max}} \frac{C_p(m,r)}{v(r)} \frac{\partial V(r)}{\partial \ln r} d\ln r \qquad (2)$$

$p=(i, l_k)=1,\ldots,N_0$

As was suggested by Dubovik et al. [5] we assume equal amount of the prolate and oblate spheroids and use the aspect ratio ε (ratio of largest to smallest size) instead of the axis ratio. The aspect ratio distribution $\frac{dn(e)}{d\ln e}$ is size independent. The distribution of spheroid particles can be described using two independent distributions and Eq.2 can be rewritten as:

$$g_p = \int_{\ln e_{\min}}^{\ln e_{\max}} \int_{\ln r_{\min}}^{\ln r_{\max}} \frac{C_p(m,r)}{v(r)} \frac{\partial V(r)}{\partial \ln r} \frac{\partial n(e)}{\partial \ln e} d\ln r d\ln e \qquad (3)$$

Assuming size independence of aspect ratio distribution , we have

$$g_p = \int_{\ln e_{\min}}^{\ln e_{\max}} \int_{\ln r_{\min}}^{\ln r_{\max}} K_p(r,m,e) \frac{\partial V(r)}{\partial \ln r} \frac{\partial n(e)}{\partial \ln e} d\ln r d\ln e \qquad (4)$$

As was shown in [5], the retrieval results are generally insensitive to the exact form of $\frac{dn(e)}{d\ln e}$. Thus, in our model we use $\frac{dn(e)}{d\ln e}$ fixed to the axis ratio distribution, providing the best fit to the detailed polarimetric laboratory measurements for desert dust samples [5]. The aspect ratio is varied in the range 1.44<ε<3.0.

For this chosen distribution $\frac{dn(e)}{d\ln e}$, the kernel function in Eq.4 can be integrated once over *lne* :

$$\int_{\ln e_{\min}}^{\ln e_{\max}} K_p(r,m,e) \frac{\partial n(e)}{\partial \ln e} d\ln e = K_p(r,m) \ . \qquad (5)$$

Then this kernel can be saved in small size look-up-tables (depending on r and n only) and used in forward model calculations as

$$g_p = \int_{\ln r_{\min}}^{\ln r_{\max}} K_p(r,m) \frac{\partial V(r)}{\partial \ln r} d\ln r \qquad (6)$$

Representing the atmospheric aerosol as a mixture of two fractions: spheres (s) and spheroids (un), and assuming that the volume fraction of spheroids η is size independent, $\frac{\partial V^{un}(r)}{\partial \ln r} = h \frac{\partial V(r)}{\partial \ln r}$, equation (6) can be rewritten:

$$g_p = \int_{\ln r_{\min}}^{\ln r_{\max}} [(1-h)K_p^s(m,r) + hK_p^{un}(m,r)] \frac{\partial V(r)}{\partial \ln r} d\ln r \qquad (7)$$

Here $K_p^s(m,r)$ and $K_p^{un}(m,r)$ are the kernel functions for spheres and spheroids. To solve Eq. (7) the distribution ∂*V*(*r*)/∂ln*r* is approximated by a linear combination of the base functions $B_j(r)$ (triangles in our case) with weight factors c_j:

$$\frac{\partial V(r)}{\partial \ln r} \approx \sum_{j=1}^{N_B} c_j B_j(r) \qquad (8)$$

Equation (9) can be rewritten in vector–matrix form:

$$\mathbf{g} = \mathbf{A}(h,m)\,\mathbf{C} \qquad (9)$$

The optical data are presented by vector $\mathbf{g} = [g_p]$, and weight factors by vector $\mathbf{C} = [c_j]$. The matrix $\mathbf{A}(h,m) = [A_{pj}(h,m)]$ consists of elements

$$A_{pj}(h,m) = \int_{r_{\min}}^{r_{\max}} [(1-h)K_p^s(m,r) + hK_p^{un}(m,r)]B_j(r)dr \qquad (10)$$

For every fixed value of *h* and *m* the system of linear equations (12) can be solved via the regularization approach using the well-known expression [6]:

$$\mathbf{C} = [\mathbf{A}^T(h,m)\mathbf{A}(h,m) + \gamma\mathbf{H}]^{-1}\,\mathbf{A}^T(h,m)\mathbf{g}, \qquad (11)$$

where **H** is the smoothing matrix of second differences [6], γ is the regularization parameter, and $\mathbf{A}^T$ is transpose of **A**. For every set of r_{min}, r_{max}, m_R, m_I, *h* the solution is found from Eq.(13). The regularization parameter is determined from minima of the modified discrepancy [3]:

$$r(g) = \frac{1}{N_0} \sum_p \left| \frac{g_p - A|f|}{g_p} \right| \qquad (12)$$

where ***A*** denotes the integral operator of the Fredholm integral equation (9). We assume that all optical data are measured with the same accuracy, thus their weighting factors in (12) are the same. The solutions f_k obtained for different combinations of r_{min}, r_{max}, m_R, m_I, η are ordered in accordance with their discrepancy r_k, from small discrepancy to largest discrepancy. As shown in our previous publication [3], the mean of the solution family obtained by averaging of solutions corresponding to small fitting discrepancy generally is close to the "true" solution. Typically we average about 1% of the total number of solutions and check that the increase of r_k does not lead to a significant change in the derived microphysical parameters. Thus if a sufficient number of independent input data is available, the main particle characteristics such as $\frac{dV(r)}{d\ln r}$, m_R, m_I, *h* can be retrieved.

To solve Eq.(11), the kernel functions $K_p^s(m,r)$, $K_p^{un}(m,r)$ must be calculated. The approach for spheroid kernels computations was suggested by Dubovik et al [5]. In order to reduce computation time, the algorithm uses precomputed look-up tables of aerosol scattering kernels. For discrete grids of r_k and e_l the Eq.6 will be rewritten as:

$$g_p = \sum_{l,k} \frac{dn(e_l)}{d\ln e} \frac{dV(r_k)}{d\ln r} K_p(m,e_l,r_k) = \sum_k \frac{dV(r_k)}{d\ln r} K_p(m,r_k) \qquad (13)$$

We use a trapezoidal approximation that assumes a linear dependence of the function $\frac{dV(r)}{d\ln r}$ between the discrete points of $\frac{dV(r_k)}{d\ln r}$, the corresponding details are given in [5,6]. In the lidar case, when backscattering of linearly polarized laser radiation is considered, the scattering matrix contains only two elements. These elements together with kernels for extinction are precomputed and used to calculate all optical data. For consistency, in our retrieval we use a look up table both for spheroids and spherical particles.

The volume distribution $\frac{dV(r)}{d\ln r}$ in our approach is approximated by superposition of triangle base functions $B_j(r)$. The number of these functions is usually 5-8<<N_r, so for implementation of look up tables in our program we need to recalculate the values of $B_j(r)$ at given points r_k. As we have already mentioned, the minimal and maximal radii of PSD should be determined in the process of retrieval, thus numerous inversion windows [$r_{\min}$, $r_{\max}$] need to be tested. However N_r=34 values of r_k are insufficient to cover all the inversion windows and we need to introduce interpolation between r_k to realize this procedure for the arbitrary values of r_{min}, r_{max}. It can be done by recalculating look-up table elements $\boldsymbol{K(m,r_k)}$ at intermediate radius r_k' inside the interval by using linear interpolation on logarithmic scale:

$$\mathbf{K}(\mathbf{m},\mathbf{r}_\mathbf{k}') \approx \mathbf{K}(\mathbf{m},\mathbf{r}_\mathbf{k}) + \frac{\mathbf{K}(\mathbf{m},\mathbf{r}_{\mathbf{k+1}}) - \mathbf{K}(\mathbf{m},\mathbf{r}_\mathbf{k})}{\ln r_{k+1} - \ln r_k}(\ln r_k' - \ln r_k) \tag{14}$$

The look-up tables used permit inversions within the interval 0.003 - 25.823 μm, and interpolation relationship (14) increases the total amount of inversion windows up to 100 which is sufficient for the retrieval algorithm presented here.

3. SIMULATION RESULTS.

To estimate the uncertainty of the retrieval of microphysical parameters with the proposed algorithm, numerous numerical simulations were performed. In simulations we used a bimodal size distribution with modal radii of fine and coarse modes r_0^f =0.1 μm and r_0^c =1.5 μm. Particle number densities in the modes were N^f=1 cm^{-3} and N^c=0.001 cm^{-3}; refractive index in both modes is m=1.50-i0.0005. The simulation was performed for the most common version of MW Raman lidar based on a tripled Nd:YAG laser. This lidar provides three backscattering coefficients at 355, 532, 1064 nm and two extinctions at 355, 532 nm wavelengths (3β+2α configuration). In the case of irregular particles the depolarization ratios δ_i at multiple wavelengths are also available.

Fig.1 shows the results of PSD retrieval for 3β+2α+2δ data set (cross-polarized components at 355 and 532 nm wavelengths are considered), the input optical data are assumed to be free of errors. The mean of the obtained family of retrieved PSD is close to the initial one; the error of total particle volume V estimation is below 3%. The retrieved refractive index is m=1.52-i0.001. When spherical kernels are used the main features of the bimodal PSD (modal radii of fine and coarse modes) are reproduced, though the error of total volume estimation increases to 22%. At the same time, the retrieved real part of refractive index m_R=1.39 is significantly underestimated. If we use spherical kernels with known a priory (fixed) *m*, the algorithm completely fails to reproduce the coarse mode.

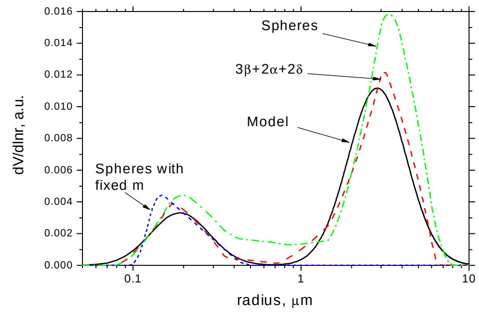

Fig.1. Model PSD (solid line) and results of retrieval for spheroids (dash) and spheres (dash-dot). Dot line presents result for spheres when exact values of refractive index is used.

The uncertainty of the retrieval strongly depends on the errors in the input optical data. The Raman technique is capable of providing aerosol backscattering and extinction coefficients with accuracy better than 10%. To estimate the accuracy of particle parameter retrievals errors in the range 0<ε<10% were introduced in the optical data in a random way. The retrieval procedure was repeated 10 times for different values of *m*. As a result we can conclude, that for 90% of the cases the errors in total volume V, effective radius r_{eff}, and mean radius r_{mean} estimation are below 30%, and errors of numerical density retrieval are below 50%. The real part of the refractive index is estimated with an accuracy better than ±0.06. the accuracy of the imaginary part estimation is approximately 50%.

For spheroidal particles such accuracy was obtained for both 3β+2α and 3β+2α+1δ (δ is taken at 355 nm) input data sets. The use of additional depolarization ratios at 532 and 1064 nm didn't lead to significant improvements in retrieval. However the situation becomes different when a mixture of spherical particles and spheroids is considered. In the absence of information about particle depolarization the spheroid volume fraction can't be retrieved with necessary accuracy. As a result the errors in the microphysical parameter retrievals are increased. Thus for processing of lidar data of irregularly shaped particles, the use of at least one depolarization ratio is essential.

4. APPLICATION OF DEVELOPED ALGORITHM TO EXPERIMENTAL DATA

The algorithm developed here was applied to the results of lidar measurements performed by the Raman lidar system *BASIL.* The lidar was operational in Achern (South Germany) between 25 May and 30 August 2007. The system measured three backscattering coefficients at 355, 532, 1064 nm wavelengths, two extinction coefficients at 355, 532 nm, and depolarization at 355 nm. On 1 and 2 August a Saharan dust outbreak episode was observed. The Lidar detected an intrusion of dust cloud, consisting of two almost separate aerosol layers: a lower layer located between 1.5 and 3.5 km and an upper layer extending between 3.0 and 6.0 km. The Particle depolarization ratio in these measurements didn't exceed 6%, thus indicating that the aerosol should contain a significant fraction of spherical particles.

The detailed analysis of the measurements will be presented in forthcoming paper. Here we have chosen the data for 2 August in height interval 3500 – 3900 m, in the center of the upper layer, to illustrate the main features of the algorithm operation. The particle size distribution $\frac{dV}{d\ln r}$ retrieved from complete input data set 3β+2α+1δ is presented in Fig.2. The coarse mode centered at 5 μm is dominating in PSD, which is typical for desert dust particles. For comparison the same picture shows PSD retrieved from data set without depolarization ratio (3β+2α) and PSD retrieved with spherical kernels. For these two cases the total volume and the real part of the refractive index is lower.

Table 1. Parameters of the particles retrieved from 3b+2a+1, 3b+2a data sets and results when only spherical kernels are applied.

	3β+2α+1δ	3β+2α	spheres
m_R	1.57±0.05	1.54	1.53
m_I	0.01±0.005	0.01	0.011
V, $\mu m^3/cm^3$	75±22	62	67
S, $\mu m^2/cm^3$	173±51	156	160
N, cm^{-3}	610±300	355	360
r_{mean}, μm	0.12±0.036	0.15	0.15
r_{eff}, μm	1.3±0.39	1.19	1.2

5. CONCLUSION

We presented a description of an algorithm for retrieval of microphysical dust properties from the results of multiwavelength lidar measurements. Simulations demonstrate that for typical dust PSD the algorithm is able to provide the estimation of aerosol microphysical parameters such as surface, volume density and particle effective radius with accuracy better than 30%. Numerical simulations demonstrate that the use of at least one cross-polarized backscatter is essential. Without the depolarization ratio we can't retrieve the volume fraction of spheroid particles in the mixture. Increasing the number of depolarization channels doesn't lead to a large improvement in the retrieval.

The application of spherical kernels can lead to the reasonable estimation of dust particle size and concentration, though the real part of refractive index is underestimated, while the imaginary one is overestimated. A lot of assumptions are used in the presented model, so additional tests, including comparison with others instruments should be performed to validate this technique. Still this first attempt to apply an algorithm incorporating spheriodal kernels to experimental data obtained with BASIL multiwavelength Raman lidar leads to reasonable results.

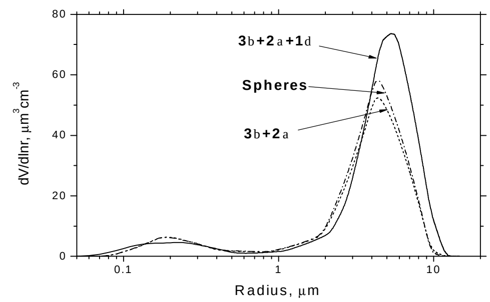

Fig.2. PSDs at 3700 m on 2 August retrieved from complete input data set (3b+2a+1d), from data set without depolarization ratio (3b+2a) and PSD retrieved with spherical kernels.

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