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Analysis of Dust Aerosol by Using Dual-Wavelength Lidar

Zhiting Wang, Lei Zhang^{*}, Xianjie Cao, Jianping Huang, Wu Zhang

Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College of Atmospheric Sciences, Lanzhou University, Lanzhou 730000, China

ABSTRACT

Simulation of dust aerosol optical property is rather difficult, due to its extremely irregular shape, which often brings about difficulties in transforming its physical properties (such as size distribution) into optical properties (such as scattering phase function) in remote sensing retrieval and atmospheric radiation model. Some recent researches reveal that homogeneous spheroids seem to be an applicable optical model when dust particles are not much bigger than the wavelength, spheroids with reasonable shape distribution can simulate the scattering phase function of dust particles quite well. Based on the existed dual-wavelength lidar inversion algorithms, a modified method is proposed in the paper. Assuming the size distributions of dust aerosol can be modeled by bimodal lognormal distributions dominated by particles ranged in coarse mode, the size distributions and lidar ratios of dust aerosol at two wavelengths can be derived from dual-wavelength lidar measurement. By applying this algorithm to the data of dual-wavelength lidar at Semi-Arid Climate and Environment Observatory of Lanzhou University (SACOL), preliminary results show that for the case of pure dust the retrieved size distribution agree with that observed by Aerodynamic Particle Sizer Spectrometer, and the derived mean lidar ratios are 45.7 ± 5.3 sr at 532 nm and 33.9 ± 1.5 sr at 1064 nm.

Keywords: Dust aerosol; Size distribution; Dual-wavelength lidar; Lidar ratio.

INTRODUCTION

The impact of mineral dust aerosol on radiation budget even climate depends mainly on particle characteristics such as size, shape and refractive index, which are highly inhomogeneous and variable. Due to the variability remote sensing is a necessary measurement for the characterization of dust aerosol. Several researches have contributed to the inversion of aerosol size distributions and refractive indexes of using the multi-wavelength lidar retrievals of extinction and backscattering coefficients (Viera et al., 1985; Donovan et al., 1997; Rajeev et al., 1998; Muller et al., 1999; Böckmann et al., 2001; Kolgotin et al., 2008). While aerosol particles are all assumed as spherical in these algorithms, it is not appropriate to apply them for the dust aerosol. Dust particles generally are with irregular shapes, and it is rather difficult deriving size distribution of dust aerosol from lidar measurement. Recent studies make progress and reveal that spheroids with certain shape distribution perform well as an optical model when dust particles are not much larger than wavelength (Mishchenko et al., 1997; Dubovik et al., 2006; Nousiainen et al., 2006, 2009, 2011). Lidar ratios

(aerosol extinction-to-backscatter ratio) at 532 nm calculated by Cattrall *et al.* (2005) based on spheroid model agree with literature values derived from observations. Results of Wiegner *et al.* (2008) reveal that calculated lidar- related parameters from spheroid model are comparable with in- situ measurements. Veselovskii *et al.* (2010) introduced spheroid model into multi-wavelength raman lidar retrieval algorithm for aerosol size distribution to account for irregularity of dust particles.

Using measurement of dual-wavelength lidar, some researchers have developed inversion algorithms for lidar ratio and other parameters showing aerosol physical properties. Dual-wavelength lidar inversion algorithm was first proposed by Sasano and Browell (1989), in which they made an assumption of constant spectral ratios of the extinction and backscatter coefficients and that lidar ratio is unknown and extinction (or backscatter) profiles and lidar ratios can be retrieved by this method. It is modified differently according to various purposes and developed into several different algorithms, in which assumption that some aerosol properties do not vary substantially in an aerosol layer are required. For example the lidar ratios, aerosol extinction-, and backscatter coefficients can be derived from dual-wavelength lidar measurements when assuming lidar ratios and spectral ratios of backscatter (Vanghan et al., 2004; Omar et al., 2007) or extinction (Lu et al., 2011) are constant in a given aerosol layer, or the spectral ratios of

^{*} Corresponding author. Tel.: +86 931 8915610

E-mail address: zhanglei@lzu.edu.cn

extinction, backscatter and lidar ratio equal some given values determined from the models of various aerosol types (Reagan *et al.*, 2004; McPherson *et al.*, 2010).

In this work we modify the Sasano-Browell method to retrieve size distribution and lidar ratio of dust aerosol through making some assumptions on the size distribution, and this method is useful for pure dust case only. Size distributions of dust aerosol are strongly dominated by large particles (Dubovik et al., 2002) and have less degree of freedom, therefore it can be approximated by bimodal lognormal distributions in which coarse modes dominate. Under the condition and assumption that composition, size distribution and shape of dust particles do not vary substantially in dust aerosol layer, and applying spheroid parameterization to model scattering properties of dust particles, the dust aerosol size distribution and lidar ratio can be derived from dual-wavelength lidar measurement. This method is described in Section 2 and in Section 3 a numerical simulation is carried out for this method. Section 4 apply this method to dual-wavelength lidar at Semi-Arid Climate and Environment Observatory of Lanzhou University (SACOL). The derived size distributions are compared with that measured by Aerodynamic Particle Sizer Spectrometer (APS). Section 5 presents the discussion and summary.

METHODS

The Physical Properties of Dust Aerosol

The dust aerosol generally distributes with bimodal mode and dominated by coarse particles, and dust aerosol size distribution can be approximated as bimodal lognormal distribution. Dubovik *et al.* (2002) showed that ratios of volume concentration in fine and coarse modes vary from 0.02 to 0.1, while Omar *et al.* (2005) reported a fine mode with volume fraction of 22%, which is likely not real. For the median radiuses of fine and coarse modes, Dubovik *et al.* (2002) presented the results of 0.12–0.15 µm (fine mode) and 1.9–2.54 µm (coarse mode), however the results of Todd *et al.* (2007) and Tanré *et al.* (2001, 2003) all showed the median radius of fine mode around 0.5 µm, in addition the median radius of coarse mode given by Tanré *et al.* (2001) vary from 1.0 to 5.0 µm.

To model dust aerosol size distribution the bimodal lognormal function is defined as follows,

$$\frac{dV(r)}{d\ln r} = \frac{C_f}{\sqrt{2\pi\sigma_f}} \exp\left[-\frac{(\ln r - \ln r_f)^2}{2\sigma_f^2}\right] + \frac{C_c}{\sqrt{2\pi\sigma_c}} \exp\left[-\frac{(\ln r - \ln r_c)^2}{2\sigma_c^2}\right]$$
(1)

where *C*- volume concentration, σ - standard deviation and *r*- median radius, and fine modes correspond to 5 values of r_f ranging from 0.1 to 0.2 µm, with $\sigma_f = 0.46$, coarse modes correspond to 15 values of r_c ranging from 1.5 to 3.5 µm, and 5 values of σ_c ranging from 0.5 to 0.7, the C_f/C_c is assumed to vary from 0.005 to 0.04 (8 values in all). The size distributions of dust aerosol surely present various

distribution shapes, however the ranges of parameters within bimodal lognormal function, especially C_f/C_c , vary not too large since only two wavelengths available, otherwise there would not be definite solution, so the retrieval is feasible only while real size distributions can be approximated by bimodal lognormal function with parameters in the value range prescribed.

Spheroids are ellipses rotated around one axis, if the axis is the longer axis, they are called prolate, otherwise oblate. Its geometry is defined by two parameters for shape and size respectively. The aspect ratio, which is defined as the ratio of the largest to the smallest particle dimension, is used to describe its shape. The size of spheroid can be described by the radius of a sphere with the same surface or volume. To simulate single-scattering properties of dust aerosol, one should use appropriate shape distribution for an ensemble of spheroids with various aspect ratios, and Nousiainen *et al.* (2006) recommended the so-called shape distribution of aspect ratio based on scattering matrix of Feldspar, which is used in this study.

Methods Description

To derive size distributions of dust aerosol from dualwavelength lidar measurement, a set of initial parameters of bimodal lognormal function- r_f , r_c , σ_c and C_f/C_c are given firstly, then lidar ratios at 532 and 1064 nm, spectral ratios of extinction and backscatter coefficients (532–1064 nm) can be calculated from the assuming size distribution using spheroids model. Giving lidar ratios, the lidar equation can be resolved using Fernald method (Fernald *et al.*, 1984) to retrieve dust extinction and backscatter profiles from lidar measurement at two wavelengths and then yield another pair of spectral ratios of extinction and backscatter coefficients. Then a quantity is defined to characterize the difference:

$$DIF = \sum_{i} \left[\frac{(\sigma_{532} / \sigma_{1064})_{i} - \sigma_{r,mod}}{\sigma_{r,mod}} \right]^{2} + \left[\frac{(\beta_{532} / \beta_{1064})_{i} - \beta_{r,mod}}{\beta_{r,mod}} \right]^{2}$$
(2)

where σ and β represent the extinction and backscatter coefficients derived from lidar measurements respectively, $\sigma_{r,mod}$ and $\beta_{r,mod}$ represent spectral ratios of extinction and backscatter coefficients calculated basing on the assuming size distribution, and summation in the Eq. (2) is for data points of lidar profile in dust layer. For each size distribution belongs to the ensemble of size distribution introduced above a difference can be calculated. All the size distributions with difference smaller than 105% of the smallest difference are averaged to produce final result.

In current method the complex refractive index is assumed to be known. In addition, the same refractive index is used at 532 and 1064 nm although in- situ measurements reveal that imaginary part of dust refractive index decreases as wavelength increases (Dubovik *et al.*, 2002; Todd *et al.*, 2007; Petzold *et al.*, 2009; Redmond *et al.*, 2010).

SIMULATION RESULTS

To test the performance of this method, numerical simulation is applied. A homogeneous dust aerosol layer of 3 km and with extinction coefficient of 0.2 1/km at 532 nm is assumed to produce lidar profile. Three model size distributions are used in all, their parameters are listed in Table 1, the refractive index used in the simulation is 1.53i0.003. Fig. 1 shows the retrieved results for different input size distributions when the refractive index is known correctly. To characterize the sensitivity of retrieval to measurement noise, the calculation above is repeated 10 times for the second model size distribution while assuming 10% measurement noise. Fig. 2 shows the calculated results. The measurement noise has an important influence on retrieval but for ground-based lidar the effect of noise usually can be neglected. Since in the current method the refractive index should be known before performing the retrieval, the sensitivity of retrieval to error in refractive index is analyzed also. Fig. 3 gives out the calculated results, in the simulation the second model distribution and refractive index of 1.53-i0.003 are used. Then various refractive indexes are used to retrieve size distribution. From the result it seems that the error in refractive index does not have definite influence on the retrieval of size distribution. But the lidar ratios from different refractive index are very different, which vary from 36.4 to 49.8 sr for 532 nm and 35.8 to 47.4 sr for 1064 nm.

APPLICATION TO OBSERVATION DATA

Data

The Dual-wavelength Mie-scattering lidar (L2S-SM II) measurements was carried out at the Semi-Arid Climate and Environment Observatory of Lanzhou University (SACOL, $35^{\circ}57$ 'N, 104°08'E, 1965.8 m) (Huang *et al.*, 2008a; Zhang *et al.*, 2010). L2S-SM II measures backscattering intensity at 532, 1064 nm and depolarization ratio profiles at 532 nm up to 24 km with a range resolution of 6 m and time resolution of 15 min, the depolarization ratio is normally used as an indicator to separate dust from other aerosol types (Murayama *et al.*, 2001). Aerodynamic Particle Sizer Spectrometer (APS-3321) provides count size distributions for particles with aerodynamic diameters from 0.5 to 20 µm, and light-scattering intensity for particles from 0.3 to 20 µm. In this paper, the data of L2S-SM II and APS during dust in March and April, 2010 are used.

Comparison between Results Retrieved from Lidar and Measured by APS

Dust events are observed at SACOL during March and April, 2010, the size distributions of dust aerosol are

 Table 1. Parameters of model size distributions used in numerical simulation.

	<i>r_c</i> /μm	$\sigma_c/\mu{ m m}$	<i>r_f</i> /μm	C_f/C_c
Model 1	1.55	0.57	0.15	0.02
Model 2	1.95	0.57	0.15	0.02
Model 3	3.40	0.57	0.15	0.02



Fig. 1. Retrievals for three model size distributions (shifted for clarity) while assuming the refractive index is known correctly. In all calculation the refractive index of 1.53-0.003i is used.



Fig. 2. Retrievals for the second model size distribution when adding 10% noise to lidar profile and assuming refractive index is known (1.53-0.003i). The retrieval is performed 10 times.



Fig. 3. Retrievals for the second model size distribution when using various refractive indexes and the refractive index of 1.53-0.003i is used for model size distribution.

retrieved from lidar measurement using the current method, and compared with that measured by APS at SACOL showing in Fig. 4. In the retrieval the refractive index of 1.53-0.001i is used. The comparison shows that for the case without definite fine mode the retrievals are reasonable except the case on March 30, 2010. However because of the difference between the size distributions and lognormal function approximation, the calculated shapes differ from the real ones, and especially the derived size distributions with lower values when radius smaller than median radius. The lidar ratios of dust aerosol can also be obtained simultaneously and Table 2 presents some results of mean values 45.7 ± 5.3 (532 nm) and 33.9 ± 1.5 sr (1064 nm), which are reasonable at 532nm and lower at 1064nm compared with literature values of 43 (532 nm) and 55 sr (1064 nm) (Liu *et al.*, 2002, 2008; Cattrall *et al.*, 2005; Liu *et al.*, 2008; Tesche *et al.*, 2008; McPherson and Reagan, 2010; Omar *et al.*, 2010), Cattrall *et al.* (2005) also derived abnormally low lidar ratio at 1064 nm based on spheroid model. The difference may arise from imperfection of



Fig. 4. Size distributions of dust aerosol retrieved from lidar measurements (solid line) and measured by APS (dot), which have been normalized by their largest values of dv/dlogr.

Date/2010	<i>r_c</i> /μm	$\sigma_c/\mu{ m m}$	<i>r</i> _f /μm	C_f / C_c	$S_{532}/{ m sr}$	$S_{1064}/{ m sr}$
17 Mar.	3.35	0.53	0.15	0.01	49.3	33.3
30 Mar.	3.05	0.59	0.16	0.04	46.8	33.6
31 Mar.	2.50	0.56	0.14	0.007	39.9	32.9
07 Apr.	3.33	0.56	0.18	0.04	52.1	33.6
11 Apr.	2.21	0.61	0.15	0.02	38.7	36.9
27 Apr.	3.24	0.56	0.15	0.01	47.2	33.3

Table 2. The derived lidar ratios and size distribution parameters of dust aerosol.

spheroid model (such as none of surface roughness in homogeneous spheroids), or from ignorance of variation of dust particles shapes along with size like Kanlder et al. (2009) showed small dust particles approach sphere more than large ones using the in-situ measurements. In this work, a numerical experiment is used to test the effect of size dependence of dust shape on lidar ratio. Two different strategies of shape distributions are applied to the size distribution assemble from AERONET site Capo verde for May to August 2010 when dust aerosol dominates. Fig. 5 shows that S_{532}/S_{1064} declines when applying shape distribution with more spheres in the range of small size. On other hand, Nousiainen et al. (2011) revealed that the optimal shape distributions of spheroid model display little similarity to those of the target particles, so the measured shape distribution, which is based on the real shape of dust particles, do not necessarily means the best ones, so currently the one inverted from scattering matrix (Dubovik et al., 2006) is used. Another possible cause is refractive index, however, for dust aerosol such knowledge are poor.

SUMMARY AND DISCUSSION

Based on the existed dual-wavelength lidar inversion algorithms, a modified method is proposed in the paper, and numerical simulation has been performed to prove its feasibility, by which dust aerosol size distributions and lidar ratios can be derived. In current method bimodal lognormal functions are used to approximate dust size distributions, and somewhat strong constraints are imposed on variation ranges of parameters in lognormal function, which is applicable for the size distributions of dust do not change largely at a given location revealed from in- situ observations, in addition the current method also need the knowledge on the refractive index of dust aerosol and an assumption of homogenous aerosol layer, as the in- situ observations show, the strong mixing action in atmospheric boundary layer, where dust aerosols occur most often, usually can make such condition be satisfied. Owing to irregularity of dust particles spheroid model is applied to simulate their optical properties.

Current method is only applicable for pure dust case. Analysis from lidar measurement at SACOL shows that the method can give out right results for size distributions without significant fine mode component. The mean lidar ratios of 45.7 ± 5.3 and 33.9 ± 1.5 sr at 532 and 1064 nm respectively are obtained, of which the value at 532 nm is acceptable and that at 1064 nm is lower. Since lidar ratio is determined by many physical properties of dust aerosol and currently our knowledge are insufficient, the interpretation to such difference needs more progresses in relevant fields.

The performance of current method depends mainly on the capability of spheroid model in the simulation of scattering properties of dust. On the whole, spheroid model works well for small dust particles and not for particles much larger than wavelength of incident light.

The linear depolarization ratio can be used to reduce the ambiguities in determining parameters of dust aerosol. However, the depolarization produced by spheroid model used here seem smaller (about 0.2 at 532 nm) compared with



Fig. 5. Frequency distributions of spectral ratios of lidar ratios S_{532}/S_{1064} calculated based on shape distribution from Dubovik *et al.* (2006) for particles lager than 0.4 µm, while for particles with size smaller than 0.4 µm using the same distribution (left) and measured distribution from Kanlder *et al.* (2009) (right), in which the size distributions are from AERONET data at site Capo_verde from May to August 2010 when dust aerosol dominates.

observation (up to 0.5, Shimizu *et al.*, 2004). This method's performance can also been improved by more powerful optical model, and nonsymmetric hexahedra seems a promising one when its database is available (Bi *et al.*, 2010). These are interesting problems for further investigations.

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