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Correlation between the lidar ratio and the Ångström exponent of various aerosol types

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A B S T R A C T

Lidar ratios and Ångström exponents of continental, maritime, and desert aerosols were calculated to evaluate the effects of aerosol composition on these parameters. Their correlation was assessed using correlation analysis and curve fitting. The Pearson correlation coefficient between the lidar ratio and the Ångström exponent was larger than 0.95 in all cases. We verified the reliability of the Pearson correlation coefficient using the significance test. The relationship between the lidar ratio and the Ångström exponent of continental aerosol can be described by a cubic polynomial model; thus, the function relation between the change in lidar ratios at different laser wavelengths depends on the fitting coefficients and the Ångström exponent. The relationship between the lidar ratio and the Ångström exponent of both maritime and desert aerosols can be described by a linear model. In these aerosols, the linear change in lidar ratios at different laser wavelengths remains unaffected by the Ångström exponent. The changes in the lidar ratio in maritime aerosol at 355 nm and 532 nm are −0.7 times and −0.18 times that at 1064 nm, respectively. For desert aerosol, the changes in the lidar ratio at 355 nm and 532 nm are 0.37 times and 1.88 times that at 1064 nm, respectively.

Introduction

Atmospheric aerosol is a complicated multiphase system, composed of the atmosphere and the particles suspended in it (Mao, Zhang, & Wang, 2002). Although only a minor constituent of the earth's atmosphere, aerosol particles have an appreciable influence on the earth's radiation budget, air quality, clouds, and precipitation, as well as the chemistry of the troposphere and stratosphere. Scattering and absorption of incoming solar radiation and long-wave terrestrial radiation by aerosol particles cause direct climate forcing, whereas indirect climate forcing can be attributed to the influence of these particles on the size distribution of cloud droplets, resulting in changes to the optical properties and lifetimes of clouds. For a better understanding of the role of atmospheric particles in climate, further investigation of the spatial and the temporal variability of their chemical and physical properties is needed (Müller, Wandinger, & Ansmann, 1999).

Lidar plays an indispensable role in aerosol detection because of its wide detection range, high spatial resolution, and real-time operation. Lidar systems are laser-based systems that operate on principles similar to that of radar (=radio detection and ranging). A pulse laser is emitted into the atmosphere. Light from the laser beam is scattered in all directions by molecules and aerosols in the atmosphere. A portion of the light is scattered back toward the lidar system. This light is collected by a telescope and focused upon a photodetector that measures the amount of back-scattered light as a function of distance from the lidar. Both the extinction coefficient and the backscattering coefficient of aerosols can be retrieved by solving the lidar equation (Song, Shi, Wang, Lu, & Hua, 2016; Yin, He, & Zhang, 2009).

The lidar ratio and the Ångström exponent also are important optical parameters of aerosols, detected by lidar. The lidar ratio is the ratio of the extinction coefficient to the backscattering coefficient of the ambient aerosols. However, given that there are two unknown parameters in the lidar equation, i.e., the extinction coefficient and the backscattering coefficient, it is necessary to set the value of the lidar ratio to solve the lidar equation using the Fernald method (Fernald, 1984; Song, Shi, Lu et al., 2016; Zhao, Hua, Mao, & Zhou, 2015). The Ångström exponent, first proposed by Ångström (1964), is closely related to the size of the aerosols. It reveals the dependence of extinction on the wavelength of the incident light (Bo, Xie, Wang, Wu, & Zhong, 2015; Song, Shi, Lu et al., 2016). Initially, the Ångström exponent was retrieved using the optical depth of the whole atmosphere, as detected by a sun photometer (Mao

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et al., 2002). More recently, the Ångström exponent of aerosols has been calculated using extinction coefficients at various wavelengths, as detected by multi-wavelength lidar (Hara et al., 2017; Tao et al., 2013).

There is close relationship between the lidar ratio and the Ångström exponent. First, both parameters are determined by the particle size distribution (PSD) and the complex refractive index. Typically, there is relationship between the lidar ratio and the Ångström exponent for specific types of aerosols (Chemyakin et al., 2016). Second, in the Fernald method, setting the lidar ratio is the basis for retrieving the extinction coefficient, while the extinction coefficient is the basis for retrieving the Ångström exponent (Ruangrungrote, Limsuwan, & Intasorn, 2013). Thus, the lidar ratio also is the basis for retrieving the Ångström exponent. Finally, it is necessary to set the value of the Ångström exponent to solve the Raman–Mie lidar equation (Mona et al., 2012).

Studying the correlation between the lidar ratio and the Ångström exponent will help resolve more about the optical properties of aerosols. To some extent, the lidar ratio reflects the directional characteristics of light scattering, while the Ångström exponent is closely related to aerosol size. Therefore, any correlation between the lidar ratio and the Ångström exponent reflects the relationship between the directional characteristics of light scattering and aerosol particle size.

Studying the correlation between the lidar ratio and the Ångström exponent also assists with retrieval of aerosol optical parameters. Any correlation between the lidar ratio and the Ångström exponent also provides a reference for setting the lidar ratio (Wu et al., 2011). If a correlation between the lidar ratio and the Ångström exponent is obtained, then an iteration algorithm can be used to retrieve the extinction coefficient, the backscattering coefficient, as well as the lidar ratio.

Although there is certainly a relationship between various aerosol types and these two parameters, aerosol type cannot be uniquely identified using only one of these parameters. However, the combination of the lidar ratio, the Ångström exponent, and the relationship between them could provide an augmented classification for aerosols.

Both the lidar ratio and Ångström exponent have been widely researched in aerosol science. An airborne high-resolution lidar was used to study aerosols in North America. According to the measured lidar ratio and backscatter color ratio, Burton et al., 2012 proposed a method for aerosols classifying. Both the lidar ratio and the backscatter ratio were used to study aerosols by Su, Liu, Wu, McCormick, and Lei (2013) in Hampton, with data obtained from a multi-wavelength Raman lidar. Optical depth, the Ångström exponent, and the PSD of aerosols during haze weather were analyzed using data from a CE318 sun photometer in Mou et al. (2014). Meanwhile, Song, Shi, Wang et al. (2016) proposed a method to retrieve the lidar ratio through iteration, in which the Mie scattering signal and sun photometer data were used.

Both parameters have been researched independently, there has been little study of their correlation. In view of this, the lidar ratio and the Ångström exponent of continental, maritime, and desert aerosols were calculated using Mie scattering theory. A correlation between the lidar ratio and the Ångström exponent was determined using a correlation analysis and curve fitting.

Theoretical basis to methodology

The components of aerosols are very complex. Different components have different complex refractive indices and PSDs (Hess, Koepke, & Schult, 1998). The most critical factor is probably the shape of the aerosol particle. In reality, aerosol particles are not typically spherical. Some aerosols even contain a large number of non-spherical particles (Kalashnikova, Garay, Martonchik, & Diner, 2013). At present, there is no definitive conclusion about the aerosol shape. Meanwhile, existing algorithms for analyzing the optical properties of non-spherical particle are restricted to limited applications and conditions (Xu, Chen, Ding, & Xia, 2014). The orientation randomness of the non-spherical particle makes this problem even more difficult to address.

Given these considerations, we assumed that aerosols were homogeneous spherical particles, and neglected the effect of multiple-scattering. Thus, the total scattering and extinction of all particles in a given volume was calculated as the sum of each particle, and the light scattering properties of a single particle was calculated using Mie theory (Xiang & He, 2007). Although using Mie scattering theory will lead to some error, simulation results can be used to evaluate optical scattering properties, providing an important reference value.

With its high power, high stability and flexible multi-wavelength selection, the Nd:YAG laser offers unmatched optical performance. Recently, the Nd:YAG laser has been used in various lidar applications. Here, we focus on the correlation between the lidar ratio and the Ångström exponent at laser wavelengths of 355, 532, and 1064 nm. The lidar ratio at wavelengths λ_1 and λ_2 is denoted as $S_{\lambda 1}$ and $S_{\lambda 2}$, while the corresponding Ångström exponent is denoted as $A_{\lambda 1,\lambda 2}$.

Microphysical properties of aerosol components

According to different sources, aerosols are divided into ten aerosol components. These are insoluble, water soluble, soot, sea salt (accumulation mode), sea salt (coarse mode), mineral (nucleation mode), mineral (accumulation mode), mineral (coarse mode), and mineral-transported particles, as well as sulfate droplets (Hess et al., 1998). To characterize size distributions, lognormal distribution are applied to each component i:

$$n_i(r) = \frac{N_i}{\sqrt{2\pi}\, r \log\sigma_i \ln 10} \exp\left[-\frac{1}{2}\left(\frac{\log r - \log r_{\mathrm{mod}\,N,i}}{\log\sigma_i}\right)^2\right], \qquad (1)$$

where $r_{\mathrm{mod}N,i}$ is the aerosol mode radius, σ_i measures the width of the distribution, and N_i is the total particle number density of component i in particles per cubic centimeter. For the dry state, the microphysical properties of aerosol components are given in Table 1.

Complex aerosols can be treated as mixtures of different components. In this case, it is assumed that there is no physical or chemical interaction among the different components. Typical compositions for continental, maritime, and desert aerosol are shown in Table 2 (Hess et al., 1998).

Definitions for the lidar ratio and Ångström exponent

Based on Mie scattering theory, both the extinction coefficient α_i and the backscattering coefficient β_i of the aerosol component i can be calculated, using the respective formulas:

$$\alpha_i = \int Q_{\mathrm{ext},i}(m, r, \lambda)\, n_i(r) \pi r^2 dr, \qquad (2)$$

$$\beta_i = \int Q_{\mathrm{b},i}(m, r, \lambda)\, n_i(r) \pi r^2 dr, \qquad (3)$$

where $Q_{\mathrm{ext},i}$ and $Q_{\mathrm{b},i}$ are the extinction efficiency factor and the backscattering efficiency factor of a single aerosol particle, respectively; and $n_i(r)$ is the PSD function.

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