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Aerosol and cloud characteristics analysis methods using multiple kinds of Raman lidar signals

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ABSTRACT

In this paper, we introduce three kinds of methods for the analysis of aerosol and cloud droplet characteristics: backscattering color ratio at two wavelengths (color ratio), aerosol liquid-water content, and cloud droplet size distribution using the liquid water and aerosol extinction coefficients. Based on theoretical perspective as well as our experimental results, we find that the liquid-water Raman scattering efficiency does not depend on four orders of particle size, but rather, depends on less than four orders, particularly in a smaller effective size distribution than Mie scattering efficiency. Therefore, we conclude that the color ratio method can be applied to aerosols, while the ratio between the liquid water Raman and extinction coefficients can be applied to cloud droplet size measurements.

1. Introduction

Aerosol characteristics are an important parameter in long-term climate, and short-term weather changes because the aerosol scattering phase function and the physics of cloud formation depend upon aerosol size, shape, and aerosol chemical-optical characteristics. Also, a variety of natural and anthropogenic aerosol sources not only endow clouds with more reflectivity, but also lengthens the lifetime of clouds by suppressing precipitation [1]. However aerosol and liquid water parameters are known only on a local basis with low time resolutions and a limited range of characteristics, because most scientists have used traditional point-measuring equipments.

Many researchers have tried to measure a variety of aerosol chemical and physical properties such as chemical components, physical characteristics, size distribution, and the refractive index by using a multi-wavelength Raman lidar and multiple-wavelength depolarization lidar. Tartarov et al. [2] provided a number of signatures of the chemical components of atmospheric aerosols using a multi-channel lidar spectrometer, but the spectrometer they used did not have a sufficient rough spectral resolution or spectral range. They also required too much time because of the low aerosol Raman scattering cross-section. Shimizu et al. [3] observed Asian dust and other aerosol characteristics using a polarization lidar. However, the depolarization method is an indirect method, and therefore, has a complex dependence upon aerosol shape and aerosol alignment in the atmosphere. Thus, this method has a limited sphere of application. Furthermore, dust is an aerosol type that can be misclassified as a cloud when the dust layer is very dense [4]. Also Asian dust events are normally transported by cold fronts, and the transported dust is frequently adjacent to, embedded in, or mixed with clouds [5].

Satellite measuring systems can give global and 24 h information about aerosol extinction or backscattering coefficients, but they have number of limitations as regards spatial and time resolutions. Because these satellite data have limitations in obtaining certain important information about aerosol chemical or physical parameters and their altitude distributions, we need to find another method that can calibrate satellite data. From meteorological and health point of view, both the aerosol parameters and spatial distribution are important, so we must measure aerosol characteristics for a large spatial distribution as well.

The concentration and profile of liquid water and water vapor can be measured by using a radiometer and a GNSS system with a low spatial resolution [6], but aerosol hygroscopic parameters cannot be measured via these methods. Cloud droplet size distribution depends upon the aerosol particle distribution and aerosol hygroscopic characteristics. In addition, aerosol atmospheric liquid water is composed of water droplets that are attached to the aerosol nucleus. To specify aerosol's hygroscopic characteristics, we must also measure these characteristics.

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Recently, other scientists have tried to combine more than two parameters in order to distinguish among aerosols by using approaches such as the lidar ratio, depolarization, and color ratio etc. [7,8].

If we can simultaneously measure a liquid water lidar signal, a nitrogen Raman lidar signal, and an aerosol Mie lidar signal, we can characterize an aerosol's hygroscopic characteristics and its cloud droplet size. The main purpose of this study is to introduce such a method of measurement and to demonstrate it in terms of aerosol hygroscopic characteristics, cloud droplet size, and particle size, using liquid water Raman, rotational, and Mie lidar systems.

2. Method

Although many scientists have tried to measure aerosols by using various lidar systems, their approaches have all treated water cloud droplets and aerosols simultaneously. However, cloud droplets have a constant refractive index and a specific size distribution that depend upon cloud height and other conditions. Here, we discuss aerosol hygroscopic characteristics, aerosol effective size distribution, and cloud droplet size distribution from a number of theoretical viewpoints.

2.1. Method for obtaining cloud droplet size distribution

Mie scattering lidar signals contain information about the aerosol backscattering and extinction coefficients, while liquid water Raman lidar signals contain information about the total liquid water volume that is attached to the aerosol. Whiteman and Melfi [9] have suggested a simple method for measuring water droplet size. They were able to measure cloud droplet size using liquid water Raman lidar signals and extinction coefficients. As calculated by Veselovskii et al. [10], the Raman scattering efficiency of liquid water is constant ($Q_{\text{Liquid Raman}} \approx 4$), and is approximately twice the extinction efficiency ($Q_{\text{ext}} \approx 2$) when the size parameter x ($2\pi r/\lambda$) is greater than 1. Hence, the total Raman signal of liquid water depends only upon the total volume of liquid water, as follows:

$$\beta^{\text{cloud}}_{\text{Liquid Raman}} = \int n(r) Q^{\text{cloud}}_{\text{Liquid Raman}}(r) \frac{4\pi r^3}{3}\, dr \approx \int n(r) \frac{16\pi r^3}{3}\, dr. \quad (1)$$

At the same time, the extinction coefficient depends upon the total area, as follows:

$$\alpha = \int_0^{\infty} n(r) Q_{\text{ext}} \pi r^2\, dr \approx \int_0^{\infty} n(r) 2\pi r^2\, dr \quad (2)$$

Because we can easily measure a cloud's optical and physical depth from the bottom of the cloud, we can easily determine a cloud's extinction coefficient [7].

Consequently, from Eqs. (1) and (2), the effective size of the water droplet can be expressed as Eq. (3).

$$r_{\text{eff}} = \frac{\int_0^{\infty} n(r) r^3\, dr}{\int_0^{\infty} n(r) r^2\, dr} = \frac{3\beta^{\text{cloud}}_{\text{Liquid Raman}}/16\pi}{\alpha/2\pi}. \quad (3)$$

where $\beta^{\text{cloud}}_{\text{Liquid Raman}}$ and α are the water droplet cloud backscattering coefficient and the extinction coefficient, respectively. These coefficients can be obtained using conventional lidar signal analysis methods [11].

Fig. 1 shows the liquid water Raman and Mie scattering efficiency of a water droplet [10]. As can be seen in the figure, when the aerosol droplet size is small, the Mie scattering efficiency is smaller than the water droplet Raman efficiency, but as the droplet size increases, the Mie scattering efficiency increases more quickly than the Raman scattering. This means that when the liquid water lidar signal is normalized by an aerosol lidar signal, the normalized liquid water Raman values are dependent upon the aerosol droplet size.

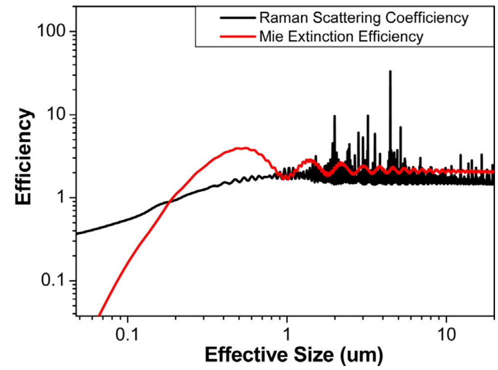

Fig. 1. Mie and liquid water Raman scattering efficient [10].

2.2. Methods for obtaining aerosol size distributions

The backscatter ratio at different frequencies–the so called "color ratio"–depends upon the size distribution, the source of the aerosol (refractive index), and the shape of the aerosol particles, and is particularly useful for inferring the mean size. Ansmann et al. [12] suggest how the color ratio, *P* and *S* may be used to infer aerosol properties, and they offer validation of their approach as obtained from aircraft flights.

Tackett et al. [13] also provide the first lidar observations of the backscatter-color ratio, and show that these quantities are directly related to aerosol properties. They show that the backscatter and color ratio are enhanced adjacent to the cloud edge, particularly near the cloud top and cloud base. Specifically, there is an increase of $31 \pm 3\%$ and $42 \pm 2\%$ in the layer-integrated median backscatter at wavelengths of 532 nm and 1064 nm, respectively, and the layer-averaged color ratio increases by $15 \pm 5\%$. These backscatter calculations suggest that our observations mode adjacent to clouds are best explained by the aerosol size distribution with a reduced number concentration, increased median radius, and decreased width, as compared to observations made at a distance from clouds.

B. de Foy et al. [14] also suggest four intensive parameters that depend only on the aerosol type and not on the concentration that can be calculated from the measurements: the lidar ratio, the aerosol depolarization ratios at 532 nm and 1064 nm, the backscatter coefficient, and the color ratio.

Recently, Burton et al. [15] and Sugimoto et al. [16] suggested that although the depolarization color ratio has not been studied extensively, it can provide information about the relative size of non-spherical particles and may also provide a means of distinguishing between smoke and pollution.

2.3. Method for obtaining aerosol hygroscopic characteristics

Normally, atmospheric aerosol contains hydrogen-bonded water, a kind of water that has a different spectrum from aerosols that contain water vapor or bulk water. If we simultaneously measure the Raman signal of this bonded water and the aerosol-scattering Mie signal, we can then extract useful information about the hygroscopic characteristics of the aerosol. For example, in a non-hygroscopic aerosol we find a strong Mie scattering signal even though we find a weak liquid water Raman lidar signal, and vice versa. When the Raman and Mie lidar receivers have the same lidar