

STUDY OF AIRBORNE PARTICULATE MATTER USING MULTIWAVELENGTH RAMAN LIDAR

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ABSTRACT

The Raman lidar system of Penn State University has a distinct advantage in measuring the optical extinction profiles at several different wavelengths simultaneously. The ratios of aerosol extinction profiles at different wavelengths are examined and model calculations are compared with the measurements. Model simulations show that the ratio of extinction is size dependent for accumulation mode particles; while for the larger size particles referred as coarse mode particles, the ratio is size independent and approaches unity. When an aerosol cloud layer is present in the laser path of Raman lidar, the analysis of the ratio of the extinction coefficient at different wavelengths can be used to describe the variations of aerosol size in the cloud layer. The extinction measurements at multiple wavelengths show unique information about particle characteristics, which can not be obtained from the single wavelength profiles.

1. INTRODUCTION

Characterization of airborne particulate matter has been a major challenge to researchers. Recent studies have associated increases in airborne particulate matter with increased morbidity and mortality, particularly in elderly and respiratory impaired individuals. Knowledge of aerosol optical properties assumes significant importance in the wake of studies strongly correlating airborne particulate matter with adverse health effects [1, 2, 3]. Along with health issues, aerosol particle distributions have significant implications for aesthetics of the natural environment and for climatic change [4, 5]. Additionally, airborne particle distributions have significant influence on the visibility that effects scheduled aircraft traffic.

Lidar (light detection and ranging) techniques have been used to make remote sensing measurements of the aerosol optical extinction and other properties associated with the particles in the atmosphere. It has been shown that the variations of optical extinction are useful in understanding the evolution of pollution events [6].

Previous efforts have developed extinction algorithms at visible wavelengths for Raman lidar [7, 8, 9, 10, 11]. The scattering cross-section for the rotational Raman signal

near the maximum of the distribution of states is almost independent of temperature, and thus it provides an excellent molecular profile to determine the optical extinction [10, 12]. The major challenge to obtain reliable aerosol extinction coefficient profiles at ultraviolet wavelengths is due to the attenuation of ozone absorption at ultraviolet wavelengths. We show that the ozone absorption coefficient profile can be calculated from the ozone density profile using the ratio of vibrational Raman shifted signals of oxygen/nitrogen (277/284 nm) from the Raman lidar measurements [6, 13].

2. METHOD

We have used Raman lidar and backscatter lidar techniques to measure the optical extinction and scattering properties as part of the NARSTO-NE-OPS (North American Research Strategy for Tropospheric Ozone - North East - Oxidant and Particle Study) during the summers of 1998, 1999 and 2001. The NARSTO-NE-OPS project is dedicated to the investigation of the sources of chemical species and particulates during atmospheric pollution episodes. The program includes the instruments that are most useful for describing the evolution of pollution events and examining the controlling influence of local meteorology on the distributions of particulate matter and chemical species in the lower atmosphere.

Lidar Atmospheric Profile Sensor (LAPS) is a Raman lidar instrument developed at Penn State University. LAPS is capable of measuring vertical profiles of water vapor, temperature, and optical extinction from scattering of the 532 nm transmitted beam. Also, profiles of water vapor, ozone, and optical extinction from scattering of the 266 nm transmitted beam are measured. Optical extinction, which is a measure of the total attenuation of a laser beam due to scattering and absorption, is obtained from analysis of the slope of the profiles of Raman scatter return signals at 607, 530 and 284 nm. The extinction is determined based upon measuring the gradient of the signal relative to the slope expected for the number density gradient of the neutral atmosphere. The neutral atmospheric density gradient can be obtained from the LAPS temperature profile and surface pressure, or by using a linear atmospheric model, which is usually accurate enough when the aerosol extinction is large [10].

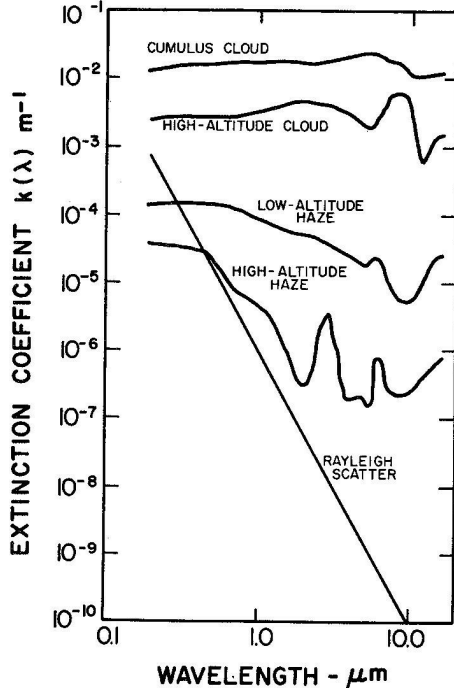


Figure 1. Aerosol extinction coefficient as a function of wavelength for different atmospheric conditions [15].

The aerosol extinction can be derived from lidar equation as [10, 12],

$$\alpha_{\lambda_R}^{aer} = \frac{\frac{d}{dz} \left[\ln \left(\frac{N(z)}{P(z)Z^2} \right) - \alpha_{\lambda_T}^{mol} - \alpha_{\lambda_R}^{mol} - \alpha_{\lambda_T}^{abs} - \alpha_{\lambda_R}^{abs} \right]}{1 + \frac{\lambda_T}{\lambda_R}} \quad (1)$$

where $\alpha_{\lambda_R}^{aer}$ is the aerosol extinction at the receiving wavelength; α_{λ}^{mol} and α_{λ}^{abs} are the molecular scattering and absorption coefficients at transmitting and receiving wavelengths. $N(z)$ represents the number density of all the molecules, z is the altitude of the scattering volume element, λ_T is the wavelength transmitted, λ_R is the wavelength received, and $P(z)$ is the power received from altitude z . Also, a telescope form factor, which is generated by the out-of-focus ray bundle overfilling the detector, has a near field effect below 800 meters. It is applied in this algorithm to aid in correcting and analyzing low altitude signals (up to an altitude 800 m) [6, 14].

An important attenuation factor at 284 nm is the ozone absorption. The ozone absorption coefficient profile can be calculated from ozone density profile which is obtained by taking the ratio of vibrational Raman shifted signals of oxygen/nitrogen (277/284 nm) from the LAPS measurements [10]. To obtain aerosol extinction at ultraviolet wavelengths, the ozone absorption was removed [6].

Figure 1 shows the expected volume extinction coefficients for several kinds of atmospheric conditions. Rayleigh scattering indicates that the scattered intensity should be inversely proportional to the fourth power of

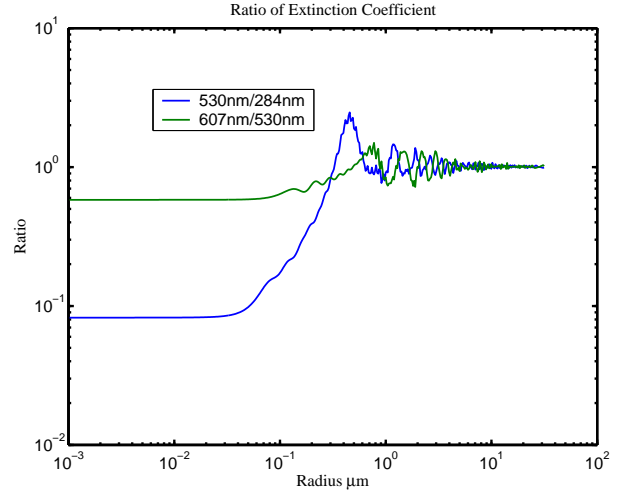


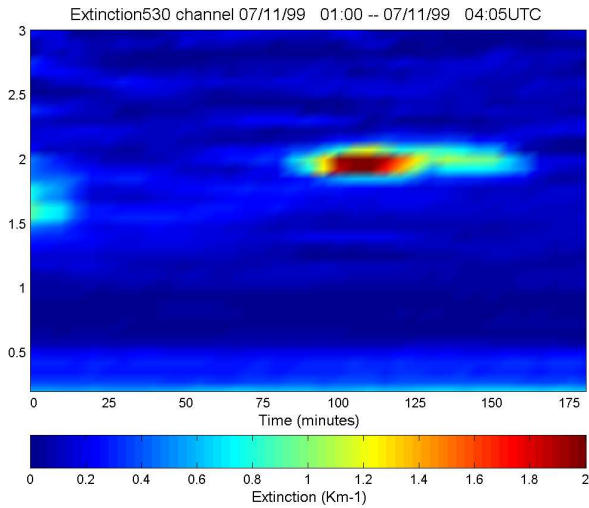
Figure 2. Ratio of extinction coefficient of 284 nm and 530 nm as a function of particle size calculated using Mie theory.

the wavelength when the particle size is small comparing to wavelength. Under haze conditions, the wavelength dependence of aerosol scattering becomes almost inversely proportional to the wavelength. While inside a cloud, the aerosol scattering is almost independent of the wavelength [15].

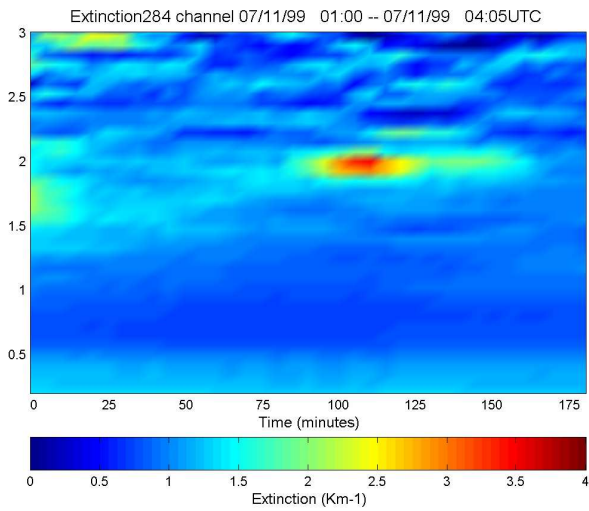
3. RESULTS

The model calculation of the ratios of extinction coefficients of 530 nm/284 nm and 607 nm/530 nm are presented in Figure 2. In our calculation, we consider only spherical particles using Mie theory. The simulations show that the ratio of the extinction coefficients of 530 nm and 284 nm are close to value 0.08, the ratio of the extinction coefficients of 607 nm and 530 nm are close to value 0.6, for fine mode particles. When the particle size is relatively small, less than about $\lambda/10$, it follows Rayleigh's theory. Both of the ratios are size dependent for accumulation mode particles where the corresponding size range is from 0.1 μm to 1 μm . For the larger size particles referred as coarse mode particle, greater than 2.5 μm , both of the ratios of the extinction coefficients are almost size independent and approach unity.

A time period was chosen as an example when there is an aerosol cloud layer passing through the laser path. The analysis of the ratio of the extinction coefficient of 530 nm/284 nm and 607 nm/530 nm shows the result relative to interpretation of the particle size information inside the cloud layer. Figures 3(a) and 3(b) show the time sequence plots of extinction at ultraviolet and visible wavelengths on the night of July 11, 1999. A cloud passed through the vertical laser beam at 2 km between 0230 UTC and 0300 UTC.



(a) Time sequence plot of extinction at 530 nm.

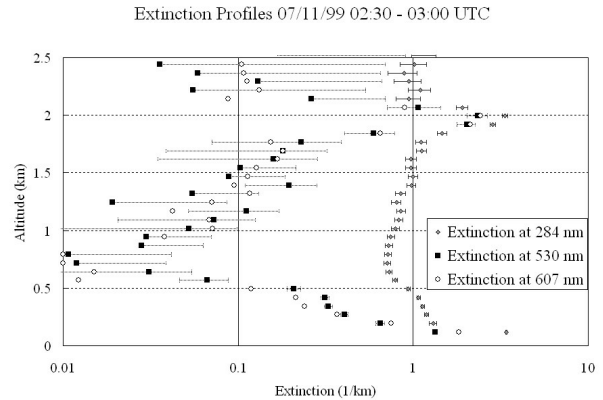


(b) Time sequence plot of extinction at 284 nm.

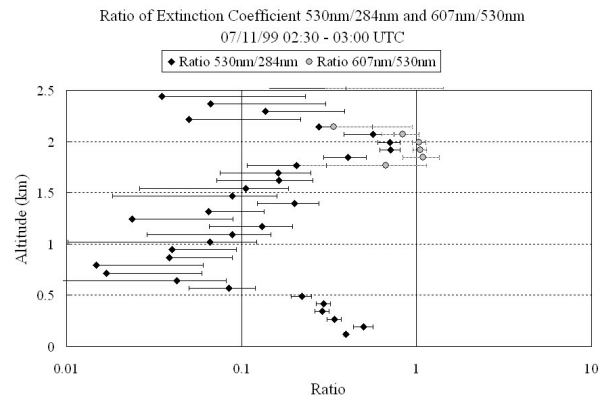
Figure 3. Time sequence plot of extinction on July 11 1999 01:00 - 04:05 UTC.

Figure 4(a) shows the 30 minute integration of extinction profiles at 607 nm, 530 nm and 284 nm during the time the cloud passing through. Note that the ozone absorption has been removed to obtain the aerosol extinction at 284 nm. The high extinction values at 2 km correspond to the cloud passing through the laser beam, which are shown in Figure 3(a) and 3(b). The error bar for the extinction at 607nm was not shown in the figure because it is relatively larger and will overlay on the other points, which makes the plot difficult to read. Figure 4(b) shows the ratios of the extinction coefficients from the results in Figure 4(a). The ratio of 530 nm and 284 nm is very close to 1 inside the cloud at 2 km altitude, which follows the expectation presented in Figure 1 and 2. It suggests that the cloud is formed by relatively large size particles ($>1 \mu\text{m}$). Also, the ratio 530 nm and 284 nm approaches 1 near the ground and indicates the higher concentration

of large aerosols at lower altitude. Because the extinction coefficients at 607 nm and 530 nm are very close to each other, and their values are relatively small ($< 0.2 \text{ km}^{-1}$) outside the cloud, as shown in Figure 4(a), there is significant amount of error when we calculate the ratio of 607 nm and 530 nm outside the cloud. Therefore, we have only included those points for the ratio of 607 nm and 530 nm that are inside the cloud layer, where they are statistical significant, see Figure 4(b). As shown, the ratio of 607 nm and 530 nm is very close to 1 inside the cloud at 2 km altitude, which also follows the expected result.



(a) Extinction profiles of 284nm and 530nm.



(b) Ratio of extinction coefficient of 530 nm and 284 nm.

Figure 4. Analysis of the ratio of extinction coefficient of 530 nm and 284 nm on July 11 1999 02:30 - 03:00 UTC.

4. CONCLUSION

LAPS Raman lidar system has the capability that it can measure optical extinction profiles at several different wavelengths simultaneously. Model simulations show that the ratio of extinction is size dependent for accumulation mode particles with size range from $0.1 \mu\text{m}$ to $1 \mu\text{m}$; while for the larger size particles referred as coarse mode particle, the ratio is size independent and approaches to

the value 1. When an aerosol cloud layer is present in the laser path of Raman lidar, the analysis of the ratio of the extinction coefficient of visible (530 nm) and ultraviolet (284 nm) wavelengths shows a striking result as the aerosol size changes in the cloud layer. This approach will be used for future investigations of the microphysical properties of clouds.

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