Ultraviolet Raman lidar for high-accuracy profiling of aerosol extinction coefficient

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An ultraviolet (UV) Raman lidar system at 354.7 nm has been developed for accurately measuring the aerosol extinction profiles. A spectroscopic filter combining a high-spectral-resolution grating with two narrowband mirrors is used to separate the vibrational Raman scattering signal of N_2 at a central wavelength of 386.7 nm and the elastic scattering signal at 354.7 nm. The aerosol extinction is derived from the Raman scattering of N_2 and the elastic scattering by the use of Raman method and Klett method, respectively. The derived results of aerosol extinction are used to compare the difference of two retrieval methods, and the preliminary experiment shows that the Raman lidar system operated in analog detection mode has the capability of measuring aerosol profiles up to a height of 3 km with a laser energy of 250 mJ and an integration time of 8 min.

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Aerosol extinction coefficient is one of the most important atmospheric optical parameters. Accurate observation of aerosol extinction is essential for forecasting of air quality and research on mechanism of Asian dust and air pollution. A single- or multi-wavelength Mie scattering lidar was reported for retrieval of this parameter[1−3]. The procedure, with all its subsequent modifications and improvements, simply suffers from the fact that two physical quantities, the aerosol backscattering coefficient and the aerosol extinction coefficient, must be determined from only one measured quantity — the elastic lidar return. The most critical input parameter in the Klett method is the lidar ratio[4]. This quantity depends on the microphysical, chemical, and morphological properties of the particles. All of these properties, in turn, depend on relative humidity. The lidar ratio can vary strongly with height, especially when marine, anthropogenic (urban, biomass burning), or desert dust particle or mixtures of these basic aerosol types are presented in layers above each other. All of these can bring much measurement uncertainty to the retrieval results of aerosol optical parameters. The Raman lidar technique has the capability of overcoming the shortages of the Mie lidar and achieving the accurate measurement of the aerosol extinction coefficient[5−12]. Because the distribution of the concentration of N_2 versus height is relatively stable in the atmosphere and the inelastic backscattering signal of N_2 is affected by aerosol extinction but not by aerosol backscattering, the aerosol extinction is determinable more accurately by the use of the intensity of the vibrational Raman scattering of N_2.

The data processing of Raman method for retrieving the aerosol extinction coefficient was described in Ref. [6] in detail. The Raman lidar equation and calculation equation for extinction coefficient are given as

$$P(\lambda_0,\lambda_{\rm N},z)=\frac{K}{z^2}Y(z)\,\sigma_{\rm N}(\pi)\,n_{\rm N}(z)\times\exp\left\{-\int_0^z\alpha(\lambda_0,z')\,{\rm d}z'\right\}\exp\left\{-\int_0^z\alpha(\lambda_{\rm N},z')\,{\rm d}z'\right\},\tag{1}$$

$$\alpha_{\rm a}(\lambda_0,z)=\frac{\frac{\rm d}{{\rm d}z}\left[\ln\frac{n_{\rm N}(z)}{z^2P(\lambda_0,\lambda_{\rm N},z)}\right]-\alpha_{\rm m}(\lambda_0,z)-\alpha_{\rm m}(\lambda_{\rm N},z)}{1+\frac{\lambda_0}{\lambda_{\rm N}}},\tag{2}$$

where K is the system constant including all depth-independent parameters; $Y(z)$ is the geometrical form factor of the Raman lidar system, in order to simplify the data processing, $Y(z)$ is not considered for long distance at which it is unity; $\sigma_{\rm N}(\pi)$ is the differential backscattering cross section of N_2, and $n_{\rm N}(z)$ is the number density of N_2; $\alpha(z)$ is the volume extinction coefficient which consists of aerosol extinction $\alpha_{\rm a}(z)$ and molecular extinction $\alpha_{\rm m}(z)$, and $\alpha_{\rm m}(z)$ can be assessed by using the standard atmosphere model. The wavelength dependence of the aerosol extinction is $\alpha_{\rm a}(\lambda_0)/\alpha_{\rm a}(\lambda_{\rm N})=\lambda_{\rm N}/\lambda_0$. Compared with the recursive formula solution of the Klett method[4], the solution of Raman method is more authentic due to its closed form, and hence the catastrophic instability phenomena cannot occur either.

The return signal is produced by Rayleigh and Raman scattering from molecules and Mie scattering from aerosol particles. Raman scattering is a weak inelastic molecular-scattering process which results in the wavelength shift between the scattered photon and the incident photon. The vibrational Raman spectrum of N_2 is shifted by 2331 $\rm cm^{-1}$ from the exciting frequency. When excited at a wavelength of 354.7 nm from a frequency-tripled Nd:YAG laser, the center of the shifted spectrum is at 386.7 nm. In order to capture the vibrational Raman signal of N_2, a high-resolution spectroscopic filter

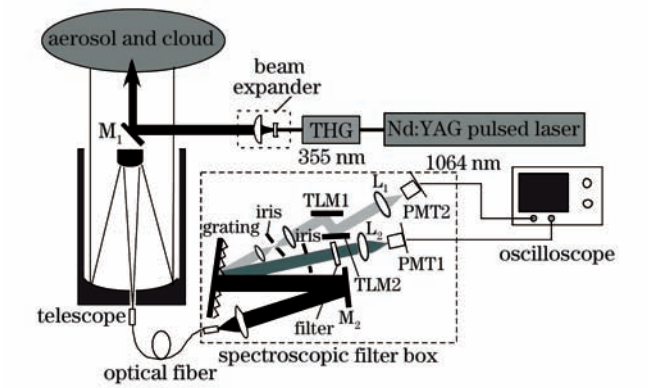

Fig. 1. Schematic of the Raman lidar system. M1,M2: mirrors; L1,L2: lenses. TLM1,TLM2: tunable laser mirrors.

is needed. A block diagram of the Raman lidar system is shown in Fig. 1. A Nd:YAG pulsed laser is employed as the light source, which produces 10-ns pulses at 20-Hz repetition rate and a maximum energy of 250 mJ per pulse at 354.7 nm after third-harmonic generation (THG). Laser outgoing is collimated by a beam expander to reduce the beam divergence and then transmitted into the atmosphere. The return lidar signal backscattered by the atmosphere is collected by a 250-mm-diameter Schmidt Cassegrain telescope and then focused into a 200-μm-core optical fiber, which sets the field of view of the receiver to 0.2 mrad considering the effect of laser beam quality factor M^2[13]. The output from the fiber is collimated and then coupled into the spectroscopic filter box, which is constructed with a high-spectral-resolution plane reflection grating mainly. The grating which has a spectral resolution of ~ 6.5 pm (15.5 GHz) instead of a narrowband interference filter diffracts the return lidar signal spatially into a Mie-Rayleigh signal, a rotational-Raman signal, a vibrational-Raman signal of O_2, N_2 and H_2O, whose center wavelengths are 375.4, 386.7, and 407.5 nm respectively. The vibrational Raman signal of N_2 is spatially separated by the high-spectral-resolution grating combined with a small pinhole, corresponding to a 2.8-nm-bandwidth filter, and a set of narrow bandwidth reflectivity mirrors with a bandwidth of 31 nm, TLM1 and TLM2. In order to compare the two retrieval methods of aerosol extinction, i.e., the Raman method and Klett method, and to verify the feasibility of the Raman lidar system, the Mie-Rayleigh channel located at the first order diffraction of the grating (354.7 nm) is used to detect the elastic scattering signal simultaneously, from which the aerosol extinction is also retrievable by the Klett method. The two scattering signals are detected by two photo-multiplier tubes (PMTs) with a pre-amplifier in analog detection mode and recorded by analog-digital conversion. The specification parameters of the Raman lidar system are given in Table 1.

The Raman lidar system has been operated for measuring the aerosol extinction coefficient at Xi'an University of Technology (108.95°N, 34.27°E), and the preliminary experiment was carried out within an integration time of 8 min (about 10000 laser shots) and a laser energy of 250 mJ. Figure 2 shows an example of preliminary measurement taken at 21:00 Chinese standard time (CST) on November 20, 2007. The received power of the Raman scattering and Mie-Rayleigh scattering signals, which

Table 1. Specification Parameters of the Raman Lidar System

Transmitter: Nd:YAG Laser	
Wavelength:	354.7 nm
Pulse Energy	250 mJ
Pulse Repetition Rate	20 Hz
Magnification of Beam Expander	5×
Beam Divergence	0.1 mrad
Receiver Optics:	Cassegrain Telescope
Telescope Diameter:	250 mm
Fiber Core Diameter	0.2 mm
Field of View	0.2 mrad
Spectroscopic Optics:	
Grating	2400 gr/mm
Bandwidth of Filter	31 nm
Detector: PMT	Hamamatsu R3896
Quantum Efficiency	23%
Gain of Pre-Amplifier	200
Bandwidth of Pre-Amplifier	12.5 MHz
Oscilloscope:	Tektronix TDS5104B
Bandwidth	1 GHz
Maximum Sampling Rate	5 GS/s

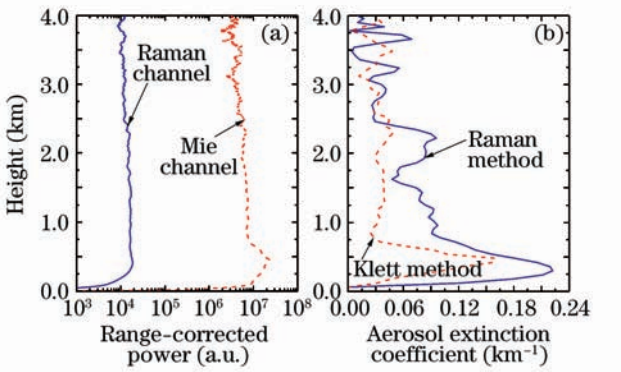

Fig. 2. Height distributions of the return signal and aerosol extinction coefficient in the lower troposphere. (a) Range-corrected lidar signals of Mie channel and Raman channel; (b) aerosol extinction coefficients which are retrieved by the Raman method and Klett method. Averaging is carried out at the laser energy of 250 mJ and an integration time of 8 min (about 10^4 laser shots) with 300-m range resolution.

are plotted as range-corrected signals versus height, are shown simultaneously in Fig. 2(a). All raw data were sampled at height intervals of 3 m and smoothed afterward with a 300-m-long sliding window. Compared with Mie channel signal, including the Mie- and Rayleigh-scattering, Raman scattering signal is lower for about three orders of magnitude and its variety trend is relatively smooth versus height due to its inelastic molecular scattering, independence of aerosol backscattering. Figure 2(b) shows the aerosol extinction profiles, which are retrieved from the Raman signal of N_2 with Raman method and from the Mie-Rayleigh signal with the Klett method, respectively. In the Raman method, all the molecular density and scattering terms are calculated from the standard atmosphere data. In the Klett

method, we made some assumptions that the height of boundary layer can be obtained as the maximum effective probing height, the aerosol extinction/backscattering ratio is 50 which means the size and chemical components of the particle cannot vary with the height, and the molecular extinction coefficient versus height is calculated from standard atmospheric model also. Comparing the shapes of the two curves of aerosol extinction coefficient, we find that the change tendency of the two curves are basically similar, especially in the height of 0.5 km where the two curves get the maximum extinction coefficient, and in the height of about 2.0 km an elevated aerosol layer exists clearly. But there is an obvious difference in the maximum value of aerosol extinctions and the shape of curves at the height range of $0.3 - 2.5$ km, and the reason might be mainly estimated due to the intrinsic problem of the Klett method as mentioned above.

Consecutive observations of aerosol extinction profile were carried out at 21:00—24:00 CST on 19 November 2007, and the observation results are shown in Fig. 3, which is plotted with 0.1-km^{-1} offset between consecutive profiles. From the results, it is clearly seen that an elevated aerosol layer exists near 2.5-km height at 21:00 CST and its height drops to 1.5 km at 24:00 CST. In order to denote the change trend of the elevated aerosol layer, we mark a dashed line manually, and it can also show the evolution of height of the atmospheric boundary layer with time roughly since above the height of the boundary layer, and the extinction value will be very small inclined to zero.

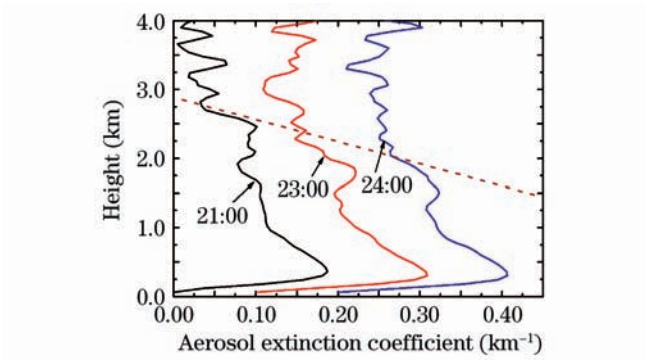

Fig. 3. Consecutive aerosol extinction profiles measured with the Raman lidar system.

In conclusion, the Raman lidar system at 354.7 nm was built for the observation of aerosol extinction profiles. A spectroscopic filter constructed with a high-spectral-resolution grating and two narrowband reflectivity mirrors was used to separate the vibrational Raman signal of N_2 spatially. The preliminary experiments showed that the Raman lidar system had the capability of accurately measuring the aerosol extinction profiles up to a height of 3 km with a laser energy of 250 mJ and an integration time of 8 min.

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