Possibilities of multi-channel lidar spectrometric technique for investigation of atmospheric aerosols and pollutions

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ABSTRACT: We present a methods and experimental data on chemical signatures characteristic for trace gases or particulate pollution (aerosol types). Measurements were made with a system that is based on a combination of a multi-channel spectrometer and a High Spectral Resolution Lidar.

INTRODUCTION

In inelastic Raman scattering the scattered signal consists of radiation that has undergone a frequency shift which is characteristic for the stationary energy states of an irradiated molecule [1]. Nowadays, Raman spectroscopy is commonly used in chemistry. Information on the radiation that results from transition between the vibrational energy states of the excited molecules, respectively, is specific to the chemical bonds and symmetry of molecules. This radiation therefore provides unique information regarding the irradiated molecule according to which the molecular species can be identified. Raman spectroscopy represents a particularly powerful tool for laser remote sensing because it allows us to both identify and quantify the trace constituent relative to the major constituents of a mixture. Raman lidar systems have become wide-spread. At the moment these systems allow for an independent quantitative measurement of the aerosol backscatter and extinction coefficient profiles on the basis of Raman scattering from nitrogen or oxygen molecules [2]. However, for a better understanding of the radiative impact of atmospheric aerosol pollution we need in future lidar measurement techniques that allow for an identification of different aerosol types, as for instance desert dust, urban haze, forest fire smoke, etc.

In this paper we present the possibilities of the multi-channel spectrometric lidar technique for investigation of atmospheric aerosols and pollutions.

METHODS AND APPARATUS

The multi-channel lidar system used in our study works on the basis of the high-spectral-resolution techniques. The system has been developed at the National Institute for Environmental Studies (NIES) [3]. The lidar system was modified with a Licel Multispectral Lidar Detector. The detector allows for the simultaneous detection of multiple spectrometer wavelengths. The spectrometer is based on a spectrograph and a Hamamatsu H7260-20 multianode, metal-channel-dynode photomultiplier with 32 photocathode elements. The detector is supplied with 32 single photon-counting systems, which provide 2-dimensional data, i.e., we obtain range-resolved signals, which in addition are separated according to the wavelength. The spectrograph is an F/3.7 Crossed Czerny-Turner ORIEL MS125 - 77400, with 120 mm focal length. For this work two different gratings were used – one is 1200 lines/mm that secure spectral resolution of about $\Delta\lambda$~6nm and a second grating with 1800 lines/mm ($\Delta\lambda$~4nm). The measurement wavelength of the HSRL was set to λ_L=532.24 nm (18 788.451 cm^{-1}). In its current design 70% of the backscattered signal are used for the multi-spectral Raman observations and 30% are used for the HSRL/polarization observations. Optional Raman edge filters were used to reduce the high intensity elastic signal that enters the multi-spectral detector. The system allows us to measure simultaneously and independently vertical profiles of Raman backscatter at 32-channels, the particle extinction coefficient, the particle backscattering coefficient, and the total (volume) depolarization ratio. The lidar ratio, the optical depth, the backscattering ratio and the (linear) particle depolarization ratio follow from these measurements.

The Raman backscatter signals obtained by the spectrometer are described by the Raman lidar equations and can be written as [2]:

$$P_{R_i}(r,\lambda_L,\lambda_{R_i}) = P_L \frac{B_R F_R(r)}{r^2} \beta_{Ri}(r,\lambda_L,\lambda_{R_i})$$
$$\times \exp\left(-\int_0^r [\alpha_p(z,\lambda_L)+\alpha_m(z,\lambda_L)+\alpha_p(z,\lambda_{R_i})+\alpha_m(z,\lambda_{R_i})]dz\right) \qquad (1),$$

where $i=1,2,...,32$ is the number of the channel, $P_{Ri}(r,\lambda_L,\lambda_{Ri})$ is the power received from distance r at the Raman wavelength λ_{Ri}, P_L and λ_L are the power and wavelength of the transmitted light, $F(r)$ is the geometrical form factor of the transmitter/receiver system, B is a constant that includes all range independent parameters, and $\beta_{Ri}(r, L, \lambda_{Ri})$ are the Raman backscatter coefficients. The extinction coefficients are denoted by α_p for aerosol particles and α_m for atmospheric gaseous molecules.

The Raman backscatter coefficients $\beta_{Ri}(r,\lambda_L,\lambda_{Ri})$ of the scatterers at all wavelengths can be determined from the received Raman scattering signal based on the knowledge of the system parameters (B and $F(r)$) and the optical depth of the atmosphere:

$$\beta_{R_i}(r,\lambda_L,\lambda_{R_i}) = \xi(r)\frac{P_{R_i}(r,\lambda_L,\lambda_{R_i})}{P_m(r,\lambda_L)} \times \beta_m(r,\lambda_L)\eta(r,\lambda_L,\lambda_{R_i}) \qquad (2)$$

Aerosol extinction (respectively optical depth) and backscatter coefficients at the laser wavelength can be obtained by simultaneous HSRL observations or Raman lidar signals obtained with the spectrometer for pure rotational Raman, signals from nitrogen molecules.

In this way, the concentration of the atmospheric components can be estimated based on the Raman backscatter coefficient for the individual wavelengths. For instance the relation of the Raman backscatter coefficient with the Raman backscatter differential cross section $d\sigma(\lambda_L,\lambda_{Ri},\pi)/d\Omega$ and the number density of molecules N_{qi} can be used for such a purpose. The relation is defined by the expression:

$$\beta_{Ri}(r,\lambda_L,\lambda_{Ri}) = N_{qi}(r)\frac{d\sigma(\lambda_L,\lambda_{Ri},\pi)}{d\Omega}. \qquad (3)$$

RESULTS AND DISCUSSION

An example of data obtained by the multi-spectral lidar detector on 6 February, 2009 is shown in Figure 1. The lidar is operated at the National Institute for Environmental Studies (NIES), Tsukuba, Japan (36.05° N, 140.12° E, 27 m above sea level). Lidar signals were recorded from 1009 to 1057 UTC with a 10 Hz repetition rate (28800 laser shots). The vertical resolution is 15 m, the spectral resolution is $\Delta\lambda$=5.7nm. The bottom panel of the figure represents the spectrum of the backscattered radiation in the vertical range from 100 m to 2000 m. In this spectrum, we can identify several peaks: the Mie (elastic) scattering at 532nm, the Raman scattering from oxygen molecules at 580nm (1556 cm^{-1}), the Raman scattering from nitrogen molecules at 607nm (2331 cm^{-1}), and the Raman scattering from water molecules at 651nm (3652 cm^{-1}). The vertical profiles of the lidar returns from these major components are presented in the upper panels of Figure 1.

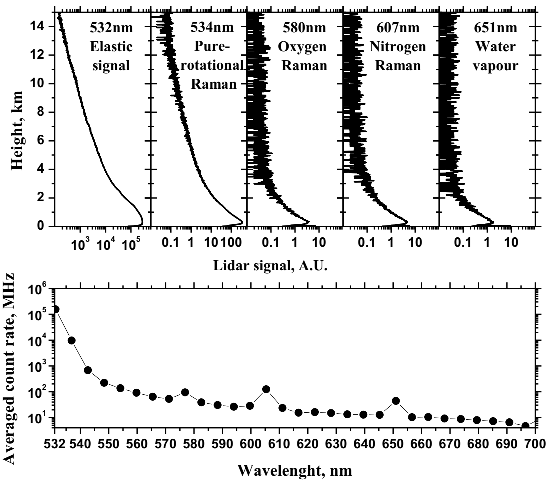

Figure1. Vertical profiles of elastic and Raman lidar signals (upper panels) and raw spectra (bottom panel) averaged across the vertical range from 100m to 2000m . Measurement time was from 1009-1057 UTC on 6 February 2009.

Figure 2 presents a measurement example of a mineral dust episode that occurred on 17 March 2009. A large mineral dust plume was transported over the lidar site on this day. We took data with our multi-spectral Raman lidar detector that was installed in the HSRL. We also carried out particle polarization measurements. The presence of the dust plume over this part of Japan was confirmed by independent measurements of the polarization lidars of the NIES Lidar Network [5].

Lidar signals were recorded from 1105 to 1745 UTC (240000 laser shots). The vertical resolution of the profiles is 150 m, the spectral resolution is $\Delta\lambda$=3.5nm. All vertical profiles of the different inelastic Raman signal components are background subtracted, range corrected and normalized to the pure-rotational Raman signal obtained at the channel with central wavelength 534 nm ($\Delta\lambda$=3.5nm). In this figure we only present the portions of the profiles of the Raman signals that are characterized by signal-to-noise ratios higher than 1, respectively. The molecular part of the optical profiles is obtained from radiosonde observations which are carried out on a routine basis at the Tateno Aerological Observatory (36.05° N, 140.13° E).

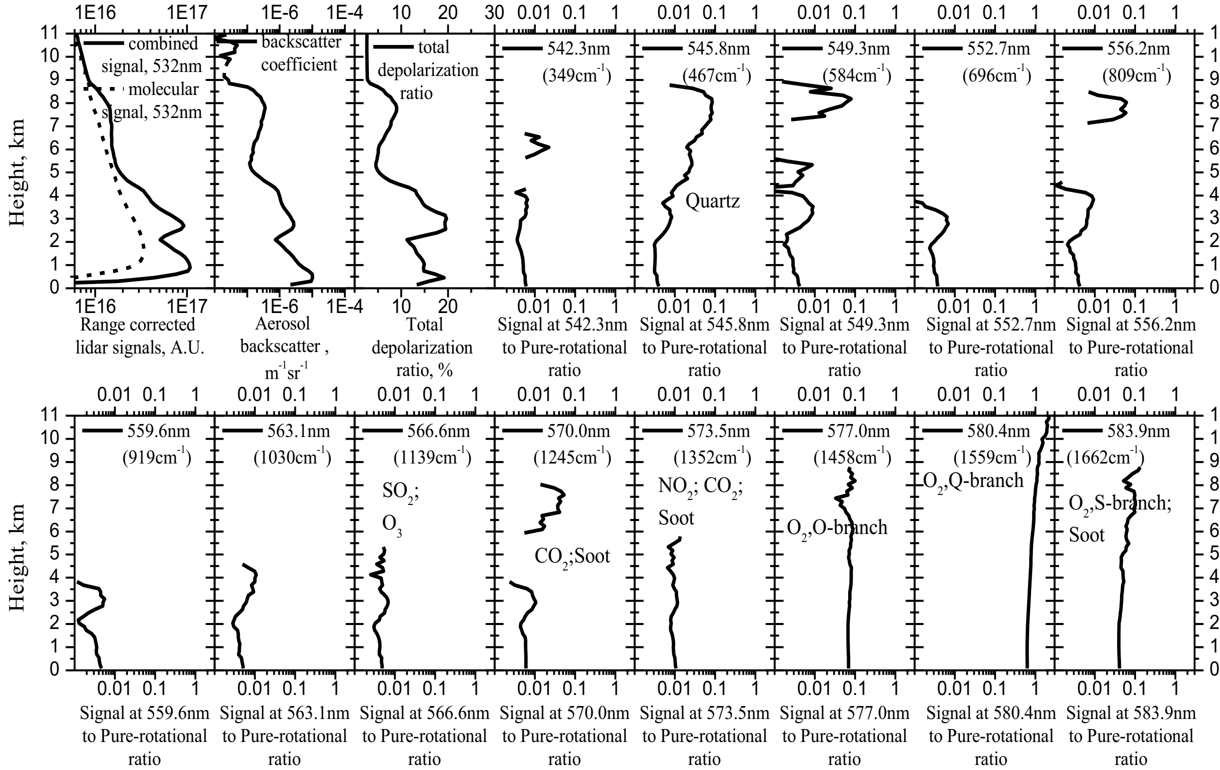

Figure 2. Vertical profiles of lidar return signals at different detection wavelengths during the advection of a mineral dust plume observed from 1105 to 1745 UTC on 17 March, 2009.

The profile of the aerosol backscatter coefficient shows the presence of several aerosol layers up to 9 km altitude above ground. The vertical profile of the total linear (volume) depolarization ratio has relatively high values in the altitude range up to 9 km, namely, about 20 % depolarization between 0.5 km and 3 km, and up to 8 % linear depolarization in heights from 3 km to 8 km. The high depolarization ratios indicate scattering from particles with non-spherical shape.

We find enhanced signal levels in the 545.8-nm Raman channel between 3 km and 9 km height. This channel corresponds to the quartz Raman line at 466 cm^{-1} [4]. Elevated values of Raman signals in that same altitude range can also be seen in the 549.3 nm channel (584 cm^{-1}). Most probably, the enhanced signal level corresponds to Raman scattering from other silicate components or specific structures (lattice) of the quartz crystals. Increased levels of the Raman signals in the height range of the dust plume (up to about 3 km) are also observed at 552.7 nm, 556.2 nm, 559.6 nm, and 570.0 nm. The signal at 566.6 nm can be related to Raman scattering from sulphur dioxide (Raman line 1151 cm^{-1}) and/or ozone (Raman line 1103 cm^{-1}). Raman scattering of soot could result in lidar return signals at 570.0 nm and 573.5 nm (Raman soot line 1331cm^{-1}) as well as at 583.9 nm (Raman soot line 1600cm^{-1}). The Raman scattering of oxygen dominates at three channels - 577.0nm (O-branch), 580.4nm (Q-branch) and 583.9nm (S-branch).

Figure 4 shows results of measurements during a mineral dust episode on 19 April 2010. The laser wavelength 355 nm was chosen as pumping wavelength for the spectrometric observations, which offered us the chance to test the spectrometer in the UV. The measurements show that the spectrometric system is capable of detecting the main atmospheric gases at 376 nm (oxygen) and 387 nm (nitrogen) as well as Raman scattering of water vapor at 407 nm.

We find a broad spectrum of lidar return signals between 400 and 540 nm. The general shape of this spectrum points to fluorescence. However, we see peaks at 470nm and 485nm on top of this broad spectrum. Most probably, these peaks can be linked to signals from different chemical components of mineral dust.

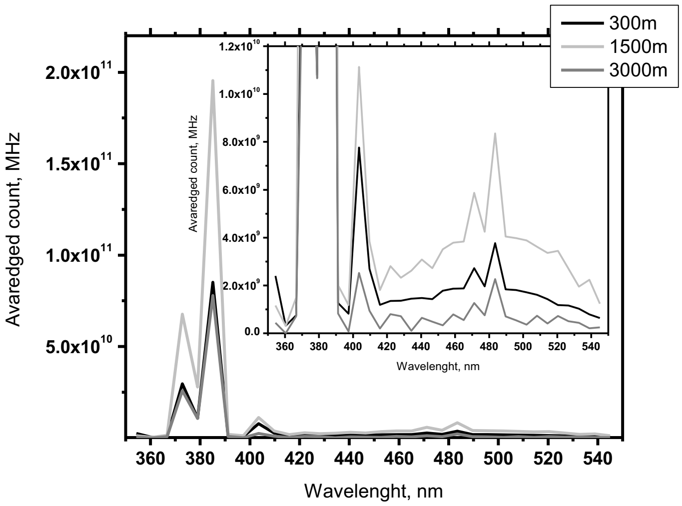

Figure 3. Raman and fluorescence spectrum at heights range 300m, 1500m and 3000 m. The measurement was carried out at Tsukuba from 15:00 to 19:00 UTC on 19 April 2010.

SUMMARY

We present first measurements of various chemical components (mineral quartz, sulphur dioxide) with a novel spectrometric technique. The measurements show that in principle it is possible to identify chemical components in particulate pollution in terms of chemical signatures contained in Raman spectra of the atmosphere. These measurements were carried out with a powerful High-Spectral-Resolution-Lidar at NIES in Tsukuba, Japan.

REFERENCES

[1] Measures, R. M., Laser Remote Sensing, 510 pp., John Wiley, New York, 1984

[2] Ansmann, A., Riebesell, M., andWeitkamp, C.: Measurement of atmospheric aerosol extinction profiles with a Raman lidar, Opt. Lett. 15, 746–748, 1990.

[3] Z. Liu, I. Matsui, and N. Sugimoto, Opt. Eng. 38, 1661, (1999)

[4] Boyan Tatarov and Nobuo Sugimoto, "Estimation of quartz concentration in the tropospheric mineral aerosols using combined Raman and high-spectral-resolution lidars," Opt. Lett. 30, 3407-3409 (2005)

[5] A. Shimizu, N. Sugimoto, I. Matsui, K. Arao, I. Uno, T. Murayama, N. Kagawa, K. Aoki, A. Uchiyama, and A.Yamazaki, J. Geophys. Res. 109, S17 (2004).