

Observation of Raman spectrums of atmospheric components by multi-channel lidar spectrometer

Boyan Tatarov, Nobuo Sugimoto, Ichiro Matsui
National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki, Japan.
e-mail: boyan.tatarov@nies.go.jp

ABSTRACT: In this paper we present 35-channel system which provides measurements of Raman spectrums of atmospheric components and aerosols. The system and the first results of the experiment are shown.

INTRODUCTION

In inelastic Raman scattering the scattered signal consists of radiation that has suffered a frequency shift that is characteristic of stationary energy states of the irradiated molecule [1]. Nowadays, Raman spectroscopy is commonly used in chemistry, since vibrational information is specific to the chemical bonds and symmetry of molecules. It therefore provides unique information by which the molecule can be identified. Raman spectroscopy represents a particularly powerful tool for laser remote sensing because it enables a trace constituent to be both identified and quantified relative to the major constituents of a mixture. In the present moment wide-spread are Raman lidar systems enable independent quantitative measurements of the aerosol backscatter and extinction coefficient profiles by obtaining Raman scattering from nitrogen or oxygen [2]. In our previous study we proposed a method to estimate dust aerosol distributions by using Raman scattering of quartz (silicon dioxide, silica), which is the major constituent of mineral dust [3]. This method combines the Raman lidar using a 466 cm^{-1} quartz line with a High-spectral-resolution lidar (HSRL) to estimate the quartz concentration in the atmospheric aerosols.

In this paper we present 35-channel system which provides measurements of Raman spectrums of atmospheric components and aerosols. The system and the first results of the experiment will be shown.

METHODS AND APPARATUS

The multi-channel lidar system has been developed based on HSRL at the National Institute for Environmental Studies (NIES)[4]. The lidar system was modified with Licel Multispectral Lidar Detector. The Licel Multispectral Lidar Detector allows simultaneous detection of multiple spectrometer wavelengths. It is based on a multianode, metal-channel-dynode photomultiplier. 32 photocathode elements together with 32 single photon counting systems provide 2-dimensional, spectral and range resolved data.

The diagram of the system is shown on Figure 1. Main characteristics of lidar in present moment are summarized in Table 1. The working wavelength $\lambda_L=532.24\text{ nm}$ ($18\,788.451\text{ cm}^{-1}$) was chosen for the HSRL measurements. In current design 70% of backscattered signal was used for multi-spectral Raman observations and 30% for HSRL/polarization observations. An optional Raman edge filter was used to reduce high intensity elastic signal into multi-spectral detector. The filter characterized by an optical density of ~ 7 for the laser line $\lambda_L=532.24\text{ nm}$ and an averaged pass band transmission of more than 93% for wavelengths from 537 to 736 nm.

Simultaneously and independently vertical profiles of Raman backscatter at 32-channels, particle extinction coefficient, particle backscattering coefficient, total depolarization ratio, can be determined. In consequence lidar ratio, optical depth, backscattering ratio and particle depolarization ratio can be obtained.

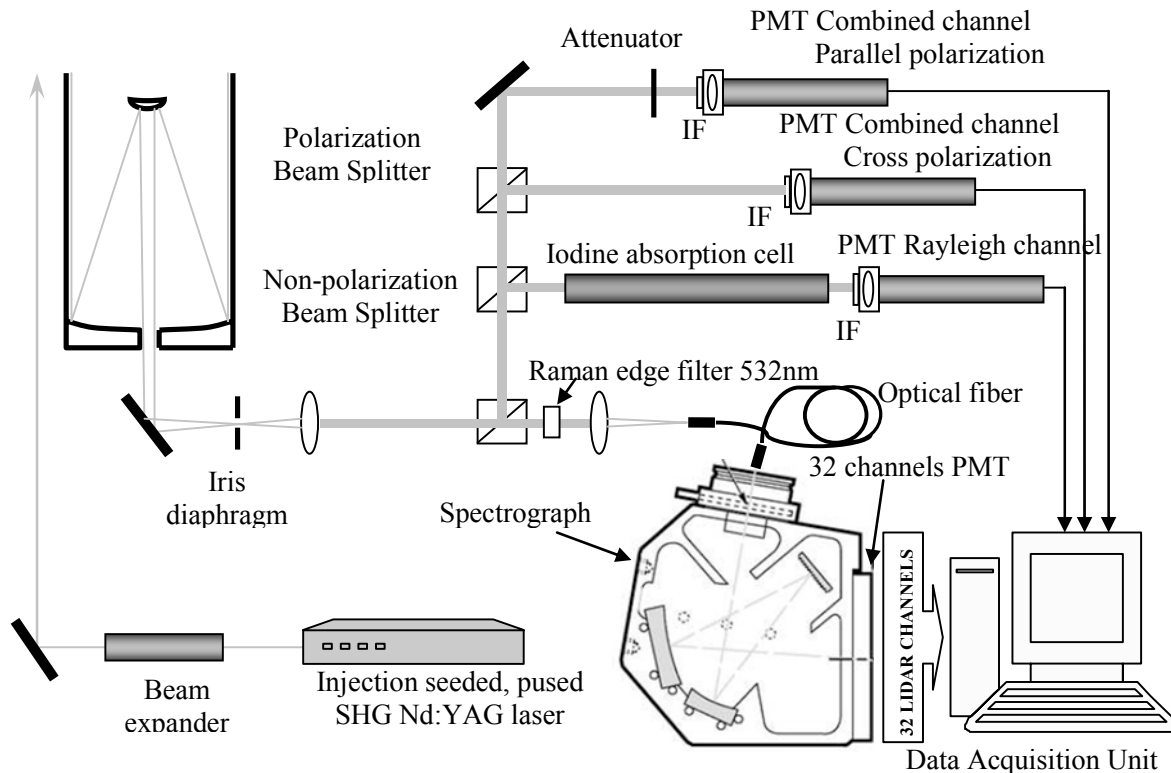


Figure 1. Scheme of the lidar system

Table 1. Basic parameters of Multi-channel lidar system

LASER SOURCE		RECEIVER	
Gain medium	Nd:YAG, Q-switched injection-seed, frequency-locked	Telescope	Cassegrainian
Polarization	linear	Main mirror diameter	560 mm
Working wavelength	532 nm	Fields of view	0.2-0.8 mrad,
Line width (FWHM)	$<0.003 \text{ cm}^{-1}$	Detections	40cm I ₂ cell, PMT R3235, bandwidth 2nm
Pulse energy	up to 400 mJ	- Rayleigh scattering	PMT R3235, bandwidth 2nm
Pulse width	$<8 \text{ ns}$	- Parallel polarization Mie and Rayleigh scattering	PMT R3235, bandwidth 2nm
Repetition rate	10-30 Hz	- Cross polarization Mie and Rayleigh scattering,	
Divergence	0.1 mrad (with 5x expander)	Spectrograph	Oriel MS125, Model 77400
DATA ACQUISITION AND PROCESSING SET		- Design	Crossed Czerny-Turner
Accusation units;	Licel transient recorder. Simultaneously record by 12 bit, 20 MHz Analog-to-Digital converters and photon-counters	- F/number	3.7
- Rayleigh and Mie channels		- Focal length	120 mm
		- Grating:	77464, Plane, Ruled, 500nm Blaze, 280-1600nm Spectral range
		- 1200 lines/mm	77421, Plane, Holographic, 500nm Blaze, 300-840nm Spectral range
		- 1800 lines/mm	Hamamatsu Multianode 32 channel array – H7260-20
- Multi-spectral channels	photon-counters only	Multi-spectral detector	
Computer	standard Linux based PC	Spectral resolution	$<6 \text{ nm}$
		- 1200 lines/mm Grating	$<4 \text{ nm}$
		- 1800 lines/mm Grating	

RESULTS AND DISCUSSION

During the experiments presented here, the lidar was placed into the National Institute for Environmental Studies (NIES), Tsukuba, Japan (36.05° N, 140.12° E, 27 m above sea level).

An example of data obtained by multi-spectral lidar detector on 6 February, 2009 is shown on Figure 2. Lidar signals were recorded from 1009 to 1057 UTC at 10Hz repetition rate (28800 laser

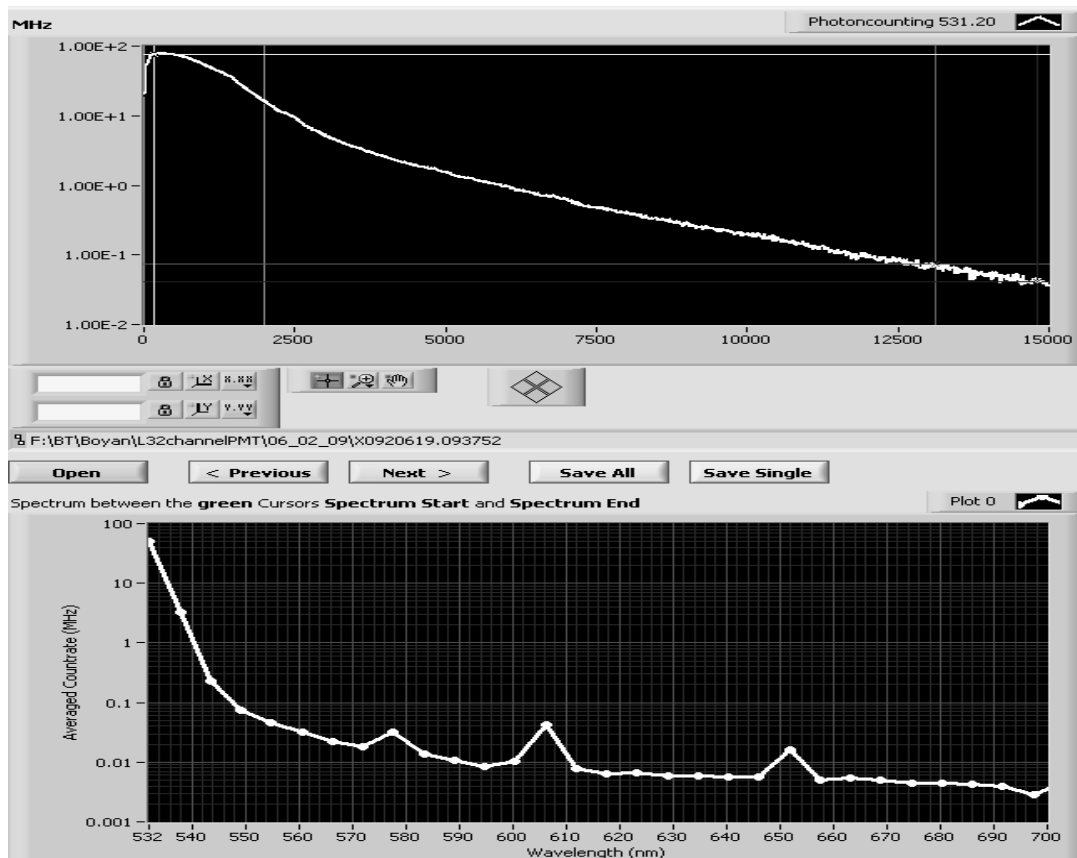


Figure 2. Vertical profile of elastic lidar signal (upper panel) and raw Raman spectrum (bottom panel) averaged over 1009-1057 UTC on 6 February 2009.

shots). The vertical resolution is 15 m, the spectral resolution is $\Delta\lambda=5.7\text{nm}$. The bottom panel on the figure represent spectrum of backscattered radiation within vertical range from 100m to 2000m. In presented spectrum one can distinguish several picks as follow: Mie (elastic) scattering at 532nm, Raman scattering from oxygen molecules at 580nm (1556 cm^{-1}), Raman scattering from nitrogen molecules at 607nm (2331 cm^{-1}), and Raman scattering from water molecules at 651nm (3652 cm^{-1}).

Figure 3 presents an example of data obtained during a mineral dust episode on 17 March 2009, by using a combination of a multi-spectral Raman lidar detector, a HSRL, and polarization measurements. Lidar signals were recorded from 1105 to 1745 UTC at 10Hz repetition rate (240000 laser shots). The vertical resolution is 150 m, the spectral resolution is $\Delta\lambda=3.5\text{nm}$. All vertical profiles on inelastic Raman components are background subtracted, range corrected and normalized on pure-rotational Raman signal obtained at 535nm. On this figure are presented only the vertical ranges of Raman signals characterized by signal-to-noise ratio higher then 1. The molecular optical profiles were obtained from the routine radiosonde observations at Tateno Aerological Observatory (36.05° N, 140.13° E). A large mineral dust plume had been transported over the lidar site on this day. The presence of the dust plume over this part of Japan was confirmed by independent measurements using polarization lidars in the NIES Lidar Network[5].

In the profiles of combined range-corrected lidar signal one can see the presence of several aerosol layers up to 9 km altitude with relatively high aerosol concentration. The vertical profile of

total depolarization ratio has relative high values in the altitudes up to 9km (about 20 % at 0.5 km and 3km, and up to 8 % at 8 km), which corresponds to scattering from particles with non-spherical shape. Relative higher values between 3km and 9km are observed at 545.8nm Raman channel which corresponding of quartz Raman line 466cm^{-1} [3]. Higher values of Raman signal at same altitudes are observed also in 549.3nm channel (584cm^{-1}). Most probably this corresponds of Raman scattering from other silicate compositions or specific structure of the quartz crystals. Relative higher values of Raman signals in the range of dust plume about 3km are observed also at 552.7nm, 556.2nm, 559.6nm, and 570.0nm. The normalized to pure-rotational Raman signal in the spectral range of oxygen scattering - 577.0nm (O-branch), 580.4nm (Q-branch) and 583.9nm (Q-branch) are almost constant with height.

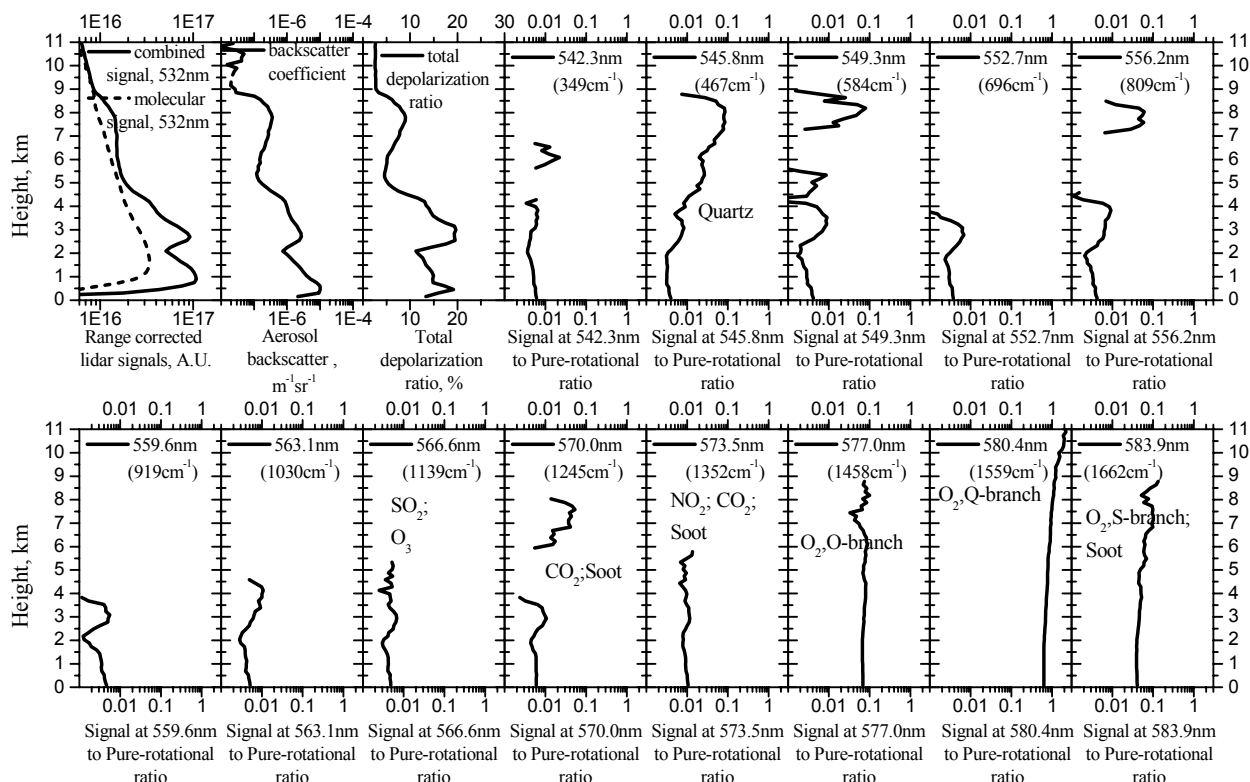


Figure 3. Vertical profiles in case of mineral dust advection obtained from 1105 to 1745 UTC on 17 March, 2009.

CONCLUSIONS

Simultaneous multi-channel Raman lidar spectrometer and HSRL measurements detected Raman spectrums of atmospheric components and aerosols and optical properties of the aerosols. The approach will be useful for characterizing and investigations of atmospheric aerosols, especially in cases of mixture of pollution gases and aerosols.

REFERENCES

- [1] Measures, R. M., Laser Remote Sensing, 510 pp., John Wiley, New York, 1984
- [2] Ansmann, A., Riebesell, M., and Weitkamp, C.: Measurement of atmospheric aerosol extinction profiles with a Raman lidar, Opt. Lett. 15, 746–748, 1990.
- [3] 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)
- [4] Z. Liu, I. Matsui, and N. Sugimoto, Opt. Eng. 38, 1661, (1999)
- [5] A. Shimizu, N. Sugimoto, I. Matsui, K. Arai, I. Uno, T. Murayama, N. Kagawa, K. Aoki, A. Uchiyama, and A. Yamazaki, J. Geophys. Res. 109, S17 (2004).