DIFFERENTIAL ABSORPTION LIDAR FOR TROPOSPHERIC OZONE MEASUREMENT USING STIMULATED RAMAN SCATTERING IN CO_2

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ABSTRACT

A UV ozone DIAL was developed which utilizes a Nd:YAG laser (266 nm output) and a single Raman cell filled with carbon dioxide (CO_2). The generated wavelengths are 276 nm, 287 nm, and 299 nm, as the first to third Stokes lines of the stimulated Raman scattering. The experimental result shows that the output energies for 276 nm, 287 nm, and 299nm were 8 mJ/pulse, 8 mJ/pulse, and 5 mJ/pulse, respectively, when the pump energy was 100 mJ/pulse and the CO_2 pressure was 0.7 MPa. As a result of some observations, it was confirmed that thirty minutes integration could observe the ozone concentration profiles up to 10 km. Some remarkable data such as stratospheric ozone intrusion, ozone enhancement in spring, urban pollution in summer, etc. are obtained using this DIAL.

1. INTRODUCTION

Our research group puts a goal on developing a set of DIAL system that can measure ozone profile from near surface up to more than 40 km and replace the existing ozonesonde. For stratospheric ozone observation the combination of a XeCl excimer laser and an Nd:YAG THG is widely used. For tropospheric ozone observation various systems are suggested. It is desirable for the design of a tropospheric ozone DIAL to be made simple and easy to handle.

A simple UV ozone DIAL has already been developed using an Nd:YAG FHG (266 nm) and a single Raman cell filled with N_2 , which produce 283 nm as the first Stokes and 303 nm as the second Stokes lines of the stimulated Raman scattering [1][2]. In this system tropospheric ozone are measured using a 283 / 303 nm wavelength pair for both the boundary layer and the free troposphere.

We developed a simple UV ozone DIAL suitable for long-term operation using carbon dioxide as a Raman active media. Main characteristics are as follows. First, this DIAL produce a wavelength pair of 276 / 287 nm less subject to aerosol interference in the boundary layer and another wavelength pair of 287 / 299 nm capable of relatively long range observation in the free troposphere. Second, it is simple and easy to handle because only a single Raman cell filled with CO_2 is needed for wavelength conversion. System description and a result of observation will be presented in the following.

2. WAVELENGTH SELECTION

Two wavelengths on the long side of the Hartley absorption band are used for tropospheric ozone measurement. Table 1 shows wavelengths obtained from the stimulated Raman scattering of the fourth harmonic of a Nd:YAG laser in various gases. The wavelength pair of 289 / 299 nm generated by the stimulated Raman scattering in deuterium and hydrogen has been utilized in many cases. The use of CO_2 enables us to produce 287 nm and 299 nm that are almost the same wavelengths as those obtained using deuterium and hydrogen. The use of O_2 is also useful for nearly the same wavelengths generation. However CO_2 would be better selection in terms of less chemical reactivity and conversion efficiency.

CO_2 can produce another wavelength of 276 nm. This wavelength is thought to be useful for more accurate measurement of ozone in the aerosol-rich boundary layer. It is estimated that the use of 276 / 287 nm wavelength pair reduces aerosol interference to less than half of that with use of 289 / 299 nm pair.

Wavelength intervals between two of any adjacent wavelengths are roughly 10 nm, which are ideal for wavelength pairs in the DIAL use. In a practical transmitter a pump beam is focused on the center of a Raman cell filled with CO_2. After the Raman cell the multi-wavelength beam produced by the stimulated Raman scattering is collimated.

Table 1. Raman active media, their Raman shifts, and the first three Stokes wavelengths of the fourth harmonic of Nd:YAG laser

Gas	Raman shift Δk (cm^{-1})	P (nm)	S_1 (nm)	S_2 (nm)	S_3 (nm)
H_2	4160	266.0	299.1	341.6	398.2
D_2	2993	266.0	289.0	316.4	349.5
CH_4	2916	266.0	288.4	314.9	346.7
N_2	2330	266.0	283.6	303.7	326.8
O_2	1550	266.0	277.5	289.9	303.6
CO_2	1385	266.0	276.2	287.2	299.1

3. SYSTEM DESCRIPTION

Fig. 1 shows the schematic diagram of the tropospheric ozone DIAL with a CO_2 Raman cell developed in the Meteorological Research Institute. The specifications are listed in Table 2. The output of an Nd:YAG laser with pulse energy of 150 mJ at 266nm is focused near the center of a 2 m Raman cell with 15 mm inside diameter using a 1.2 m focal length lens. The output from the Raman cell is collimated with a 4 m focal length lens to make the beam divergence small.

The developed lidar has two receiving telescopes. A 25 cm diameter Newtonian telescope is employed for boundary layer measurements up to about 3 km, which has two receiving channels for 276 nm and 287 nm. Another 60 cm diameter Nasmyth-Coude telescope is used for measurements in the free troposphere, which has two channels for 287 nm and 299 nm. For each receiver system, lidar return is focused at the field stop to form the field of view of 1.0 mrad, separated into two channels using a half mirror, limited to respective wavelengths with 2 nm band width interference filters, and detected by photomultiplier tubes.

Detected signals are processed by means of both 12-bit analog and 250 MHz photon-counting methods in the transient recorders. LabVIEW software is used for data acquisition and control.

Transmitted light travels coaxially with the 25 cm telescope axis. This makes the overlap height of the transmitted beam and the receiver field of view lower to about 200 m. We could make use of 266 nm for lower tropospheric ozone observation. However it is not used in this system because it would not give considerable advantages. In addition to ozone, aerosols could also be observed through a little improvement.

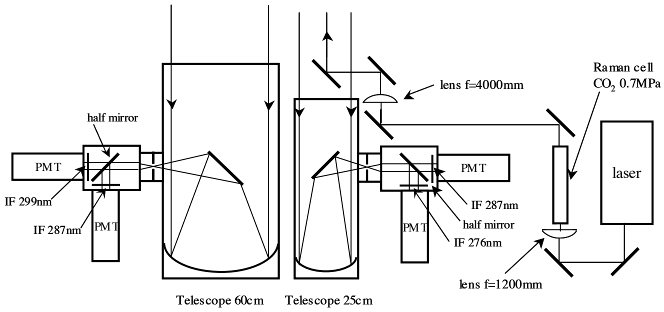

Fig. 1. Schematic diagram of the MRI UV ozone DIAL.

Table 2. Specifications

Laser		
Pump laser and its wavelength (nm)	Nd:YAG FHG (266nm)	
Pulse energy	150mJ/pulse	
Pulse repetition rate	10Hz	
Pulse duration	4～6ns	
Raman cell		
Cell length	2m	
Raman active gas and shifted wavelengths	CO_2 (S1:276nm, S2:287nm, S3:299nm)	
Beam diameter	4cm	
Beam divergence	～0.1mrad	
Telescope		
Type	Newtonian	Nasmyth-Coude
Telescope diameter	25cm	60cm
Field of view	1.0mrad	1.0mrad
Detection and processing		
Wavelengths	276nm, 287nm	287nm, 299nm
Band width (FWHM)	2nm	2nm
Detector	PMT	PMT
Signal processing	analog / photon counting	analog / photon counting

4. EXPERIMENT

Fig.2 shows the pulse energy for each wavelength as a function of CO_2 pressure in the Raman cell when the pump energy is 100 mJ/pulse. The 299 nm wavelength has the smallest energy through CO_2 pressure range. From this result the CO_2 pressure was determined to be 0.7 MPa in order to maximize the energy of 299 nm. In this condition the resulting pulse energies for 276 nm, 287 nm, and 299 nm are 8 mJ/pulse, 8 mJ/pulse, and 5 mJ/pulse, respectively. As a result of the simulation using the parameters in Table 2, observable height ranges are estimated to be about 0.2-3 km using the receiver for the boundary layer measurement and about 2-10 km using the other receiver.

The effect of the buffer gas in CO_2 is also examined. In the experiment, argon is employed for the buffer gas and the CO_2 pressure is fixed at 0.61 MPa. The result is shown in Fig. 3. The output pulse energy of the third Stokes line 299 nm decreases monotonically with increase in argon pressure, while those of the first and second Stokes lines are approximately constant. This result suggests that the buffer gas is not effective to increase the energy of the third Stokes line in the case of CO_2 use.

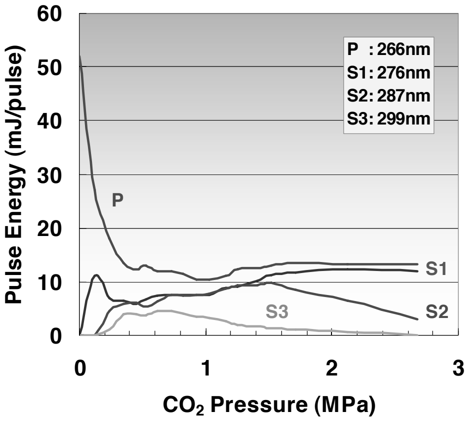

Fig. 2. The output energy obtained from the SRS of the Nd:YAG 4th harmonic as a function of CO_2 pressure.

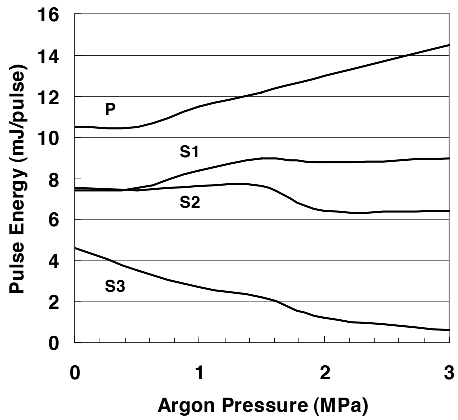

Fig. 3. The output energy obtained from the SRS of the Nd:YAG 4th harmonic in CO_2 as a function of Ar pressure.

5. OBSERVATION RESULTS

In order to verify the effectiveness of this DIAL, tropospheric ozone observations have been conducted at the Meteorological Research Institute in Tsukuba since November 2004. As an example, the results obtained on February 2-4, 2005 will be presented. The 57-hour observation was started at 0000UTC on February 2, 2005. On the way it was interrupted for 2 hours around 2200UTC on February 2 due to system maintenance.

In this example, raw data were obtained with the laser pulse energy of 82 mJ/pulse (266nm), range gate of 7.5 m, and integration time of 5 minutes. After data acquisition, post data processing was carried out as follows. The raw data were integrated every 13 bins in the range, thus the range gate becomes 97.5 m. Subsequently, six temporal data are added together to obtain a profile every 30 minutes. Then analog and photon counting returns are connected for each wavelength to obtain the ozone profiles for both the boundary layer and the free troposphere, where the third order spline fittings are performed using nine data points in the range. Finally, both profiles are connected each other. Correction for molecular extinction is carried out, while those for aerosol extinction and backscattering, and for absorptions by other gases are not.

Fig. 4 shows the time-height cross-section of tropospheric ozone from 0000UTC on February 2 to 0900UTC on February 4. Lower stratospheric ozone and its variation, two stratospheric ozone intrusions started at 0000UTC on February 2 and 1200UTC on February 3, low ozone density in the free troposphere after the intrusions, and relatively dense ozone in the boundary layer are depicted in the figure.

Both ozone intrusions have different descending rate and finally reach the height around 2 km. This height is above the top of the boundary layer. Therefore the ozone seems not to have intruded into the boundary layer.

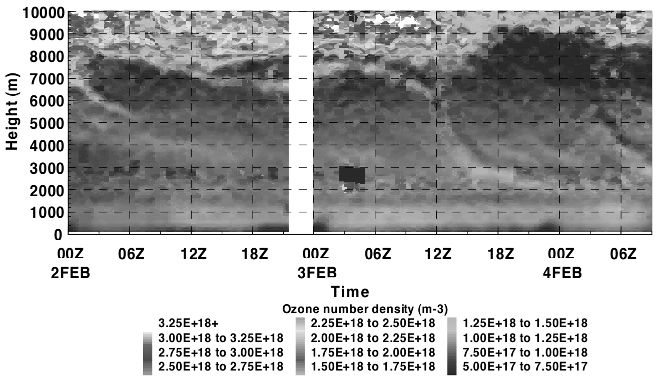

Fig. 4. Time-height cross-section of tropospheric ozone over Tsukuba on February 2-4, 2005.

6. DISCUSSION

Meteorological analysis was carried out using the observed ozone profiles, objective analysis made by the Japan Meteorological Agency, soundings, and an ozone profile obtained from the ozonesonde observation.

The ozonesonde data suggested that the stratospheric ozone depicted around 9 km in Fig. 4 was identified as that just above the first tropopause. Temporal variation of soundings indicated the descent of the upper front corresponding to the first intrusion of ozone. This implies that the first intrusion is related to the tropopause folding. Fig. 5 shows a time-height cross-section of potential vorticity and potential temperature over Tsukuba. This figure is similar to Fig. 4, implying that the isentropic potential vorticity analysis is effective for this case. As an example, isentropic potential vorticity, geopotential height, and wind vector distributions on the 285 K isentropic surface are illustrated in Fig. 6. These analyses suggested that the first descent was also related to the cut-off low and the second was affected by enhancement of baroclinicity associated with the warm air advection from the southwest. Analysis for the observed ozone in the boundary layer showed diurnal variation after breaking of the temperature inversion, probably due to photochemical reaction. Other data with respect to urban air pollution are presented in this volume [3]. Comparison with ozonesonde is also described in another manuscript in this volume [4].

As described above, the developed DIAL is useful for various meteorological applications such as STE, ozone enhancement in spring, urban air pollution in summer, etc.

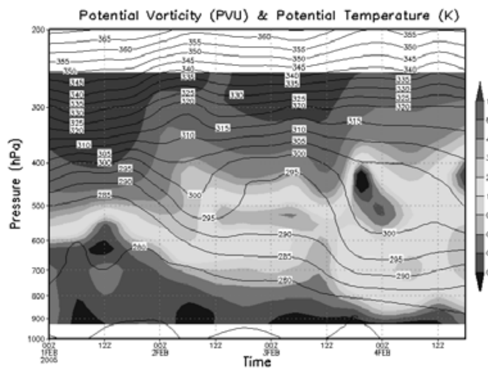

Fig. 5. Time-height cross-section of potential vorticity and potential temperature over Tsukuba on February 1-4, 2005.

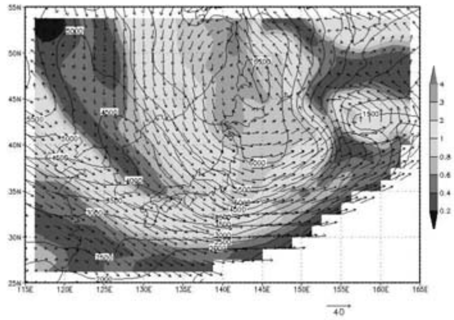

Fig. 6. Isentropic potential vorticity, geopotential height, and wind vector distributions on the 285 K isentropic surface on February 2, 2005.

7. CONCLUDING REMARKS

A UV ozone DIAL was developed which utilizes a Nd:YAG laser and a single Raman cell filled with carbon dioxide (CO_2). The generated wavelengths are 276 nm, 287 nm, and 299 nm, as the first to third Stokes lines of the stimulated Raman scattering. It was confirmed that thirty minutes integration could observe the ozone concentration profiles up to 10 km. Some remarkable data are obtained using this DIAL, suggesting that the DIAL is useful for various atmospheric researches.

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