A new differential absorption lidar for NO_2 measurements using Raman-shifted technique

Shunxing Hu (胡顺星), Huanling Hu (胡欢陵), Yinchao Zhang (张寅超), Jun Zhou (周 军), Guming Yue (岳古明), Kun Tan (谭 琨), Yufeng Ji (纪玉峰), and Ben Xu (徐 赟)

Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Hefei 230031

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Based on Raman-shifted wavelengths of D_2 and CH_4 pumped by third harmonic of Nd:YAG laser, a differential absorption lidar was presented in this paper and had been constructed for probing environmental NO_2 concentration. NO_2 experimental measurements were carried out at Anhui Institute of Optics and Fine Mechanics in Hefei. Some NO_2 measurement results were given and discussed.

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Differential absorption lidar (DIAL) is a powerful technique and has great potential in remote sensing of pollutant gases, such as O_3, NO_2, SO_2 and so on[1,2]. Several kinds of light source, such as solid-state tunable lasers, dye tunable lasers, were adopted to measure NO_2 in atmosphere. But they are restricted to actual applications because of their expensive or inconvenient in operation and maintenance. Raman cell, as a kind of wavelength shifter, exhibits its benefits of high efficiency and simplicity of the setup[3]. This Raman wave-shifter is in common use in tropospheric ozone DIAL systems[4,5]. In this paper we present a new method for NO_2 concentration measurements in the planetary boundary layer (PBL). This method is based on Raman-shifted wavelengths of D_2 and CH_4 pumped by the third harmonic of Nd:YAG laser.

Two pulsed laser beams are emitted alternately at the same path into the atmosphere, one of which is strongly absorbed by NO_2 and the other is not or slightly. We present two wavelengths of 395.60 and 396.82 nm to measure NO_2 concentrations. The laser beam at wavelength of 395.60 nm, as a strong absorption line ($\lambda_{\rm on}$) shown in Fig. 1, is the Raman-shifted wavelength of CH_4 gas pumped by the third harmonic of Nd:YAG laser. And another at wavelength of 396.82 nm, as weak absorption line ($\lambda_{\rm off}$), is the Raman-shifted wavelength of D_2 gas pumped by the third harmonic of Nd:YAG laser. The backscattered light signals due to air molecules and aerosols are collected with a telescope. Then the profiles of NO_2 can be derived according to the conventional DIAL equation from the two wavelengths return signals. Thus the expression of NO_2 concentrations[6] can be given as

$$N(z) = \frac{1}{2[\delta_{\rm on}(T) - \delta_{\rm off}(t)]} \times \left\{ \frac{\rm d}{{\rm d}z} \left[-\ln \frac{P_{\rm on}(z)}{P_{\rm off}(z)} \right] + B_A + E_A + E_M \right\}, \quad (1)$$

where $P_{\rm on}(z)$ and $P_{\rm off}(z)$ are the photon numbers, corrected terms of B_A and E_A are due to aerosol backscattering and extinction at the height of z. They are neglected generally. Term E_M is due to air molecular extinction at the height of z and can be corrected with radiosonde data or atmospheric molecular model. $\delta(T)$ is the temperature dependent absorption cross-section of NO_2, T is the temperature. According to Bogumil's work on NO_2 absorption cross section, the differential NO_2 absorption cross section $[\delta_{\rm on}(T) - \delta_{\rm off}(T)]$ varies small under pressure 10^5 Pa[7]. The capital letter $N(z)$ is the number density of NO_2 molecules.

The NO_2 DIAL system used for experimental measurements is mainly composed of three parts: transmitter, receiver and return signal acquisition, and controlling. Schematic of the DIAL system is shown in Fig. 2.

The transmitter includes two Nd:YAG lasers, laser trigger, second harmonic crystals, third harmonic crystals,

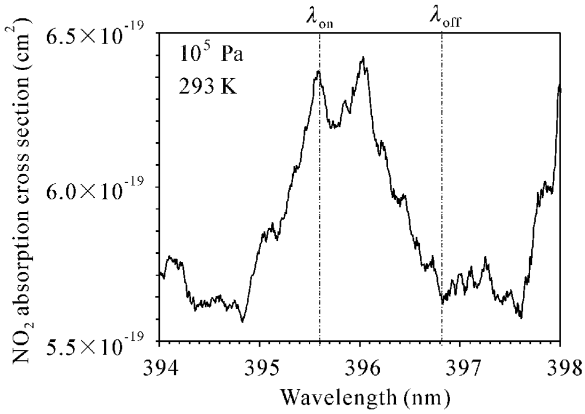

Fig. 1. NO_2 absorption spectrum under temperature of 293 K and pressure of 10^5 Pa.

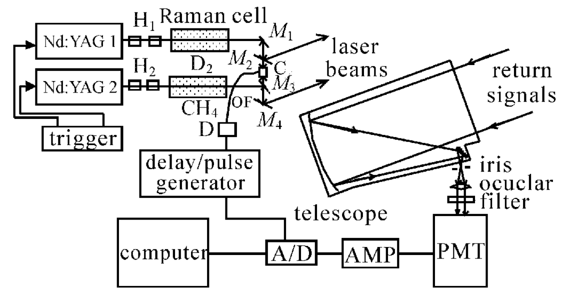

Fig. 2. Schematic of NO_2 DIAL system. H_1, H_2: second harmonic crystals and third harmonic crystals; $M_1 - M_4$: steering mirrors; C: light collector; D: photoelectric dioxide; OF: optical fiber.

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D_2 Raman cell, CH_4 Raman cell and some mirrors. Laser beams at wavelength of 354.7 nm, generated by second harmonic crystal and third harmonic crystal, are used to pumping D_2 and CH_4 Raman cell, then Raman shifted wavelength of 395.60 nm is got from CH_4 Raman cell and Raman-shifted wavelength of 396.82 nm is obtained from D_2 gas Raman cell. The mirrors are employed to exclude unnecessary laser beams and conduct the needed lasers of 395.60 and 396.82 nm into the atmosphere.

The receiver comprises receiving telescope, iris, ocular and optical filters. The telescope is at an elevation of 30 degree to collect the return signals. The iris is adjustable to restrict the field of view (FOV) of the telescope. The ocular turns the divergence light into parallel light. The optical filter (Barr Co., central wavelength 396 nm, bandwidth 20 nm) is set to block the background light onto the surface of photomultiplier tube (PMT).

The signal acquisition and control part includes PMT, high voltage power supply (PS325/2500V-25W), amplifier, A/D card (PCL9812), time delayer and pulse generator (DG535), light collector, cooler (FACT50) and industrial computer. The PMT (9814QB) works under temperature −20 degree to reduce PMT thermal noise. Time delayer and pulse generator synchronize the step of acquisition of A/D card and emission of laser beam by controlling the start and end time of the A/D card.

A laser pulse trigger sends rectangle pulses alternately to the inputs of two Nd:YAG lasers at repetition of 20 Hz and lasers start to work. Mirrors M_2 and M_3 coated with medium film reflect laser beams of 395.60 and 396.82 nm, and permeate other laser beams. The permeating lights are collected and focused into an optical fiber, then directed onto a photoelectric diode. The diode outputs pulses to the time delayer and pulse generator, the generator outputs rectangle pulses to control acquisition time of A/D card. Return signals from PMT are amplified and acquired by A/D card and stored in a hard disk of computer for further analysis.

Before measurements, it is needed to adjust the two laser beams of 395.60 and 396.82 nm parallel to the light axis of the telescope. Both of their ernergy are about 10 mJ. As shown in Fig. 3, we adjust firstly the mirrors S_1 and S_3 with an angle reflector R and make laser beam 396.82 nm parallel to light axis of telescope. Secondly we set a plane mirror M and a fluorescence printing paper iris D_1 with two pinholes on the path of laser beam, then adjust the plane mirror and make the laser beam of 396.82 nm return back along its path and fasten the plane mirror. Finally we adjust mirror S_2 to make beam of 395.60 nm parallel to beam of 396.82 nm. Thus we assure the both laser beams parallel to the light axis of telescope and move off plane mirror M and iris D_1.

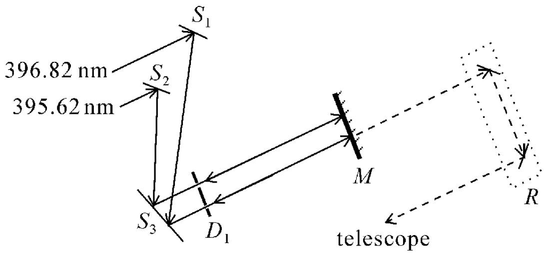

Fig. 3. Sketch map of adjustment.

The two Raman scattering laser beams of 396.82 and 395.60 nm are 10 centimeter apart from receiving telescope and their divergences are big. So this iris for restricting receiving FOV is tuned widely to minimize the lidar blind and cross range when probing. Considering of the wide FOV and wide band of the interference filter, we measured NO_2 concentrations at night. The pulse number and bin time for acquisition are preset to 40 thousand pulses and 15 m, respectively. This means that spatial resolution is 15 m. Range of signal acquisition spans 15.36 km for 1024 sampling point. The two lasers are triggered alternately. Laser repetition is 10 Hz. It takes about 34 minutes to obtain a NO_2 profile.

The NO_2 measurements have been carried out for several days after the NO_2 DIAL system operation. As shown in Fig. 4, four night measurement results were given with spatial resolution of 120 m. Left ordinate, right ordinate and horizontal ordinate represent slant range (elevation of telescope is 30 degree), altitude and NO_2 concentration, respectively. Commonly NO_2 concentration is about 20 ppb at the NO_2 DIAL system located site. Sometimes it reaches 50 – 60 ppb. In Fig. 4, symbols of "★" and "□" show the average NO_2 concentration at height 14 m above the earth surface at the DIAL site. Those NO_2 concentrations were measured with NO_2 analyzer provided by Hefei environment office. Table 1 lists the results measured with NO_2 analyzer and NO_2 DIAL system at minimum slant height 0.42 km. This table demonstrates the capability of the NO_2 DIAL system.

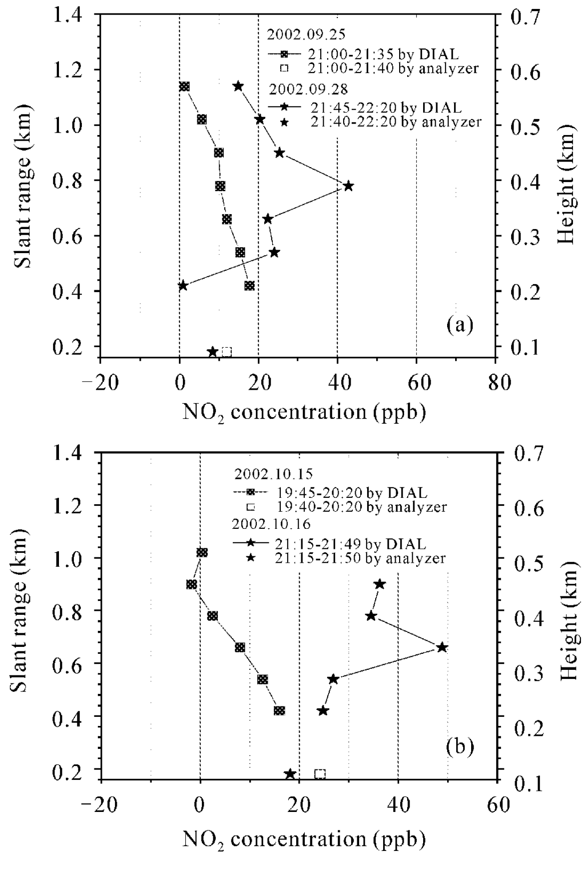

Fig. 4. NO_2 concentrations measured by DIAL and analyzer.

Table 1. NO_2 Concentration Measured with Analyzer at Height of 14 m and NO_2 DIAL System at Slant Height of 0.42 km (Concentration Unit: ppb)

Date		9.25	9.28	10.15	10.16
Analyzer	NO_2	12.04	8.34	24.2	18.2
	STD	2.0	5.1	5.3	6.1
DIAL	NO_2	17.7	1.0	15.96	24
	STD	5.1	5.5	6.0	6.4

The differential NO_2 concentrations between DIAL and analyzer are better than 10 ppb.

We present a new DIAL method for profiling NO_2 concentration with Raman-shifted wavelengths of D_2 and CH_4 pumped by third harmonic of Nd:YAG laser. NO_2 concentrations were measured with NO_2 DIAL system at slant range from 0.4 to 1.2 km in PBL. The primary measurement results demonstrate the measurement capability of the NO_2 DIAL system within error of 10 ppb.

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