

P2-3 Development of a Multiwavelength DIAL System and Ozone Measurement in the lower Troposphere

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I. Introduction

The chemical reaction of atmospheric trace substances such as Ozone and NO_x has been reported to cause urban pollution [1]. The vertical concentration profiles of these trace substances, which are needed to validate urban pollution models, can be conveniently measured by a ground-based differential absorption lidar (DIAL). In addition, long-range transport models to estimate sulfur deposition in Japan and East Asia have recently been developed [2, 3], and DIAL measurement of sulfur compounds such as SO_2 are needed for validation [4].

We have developed a lidar system which is capable of precise measurement of a single species by multiwavelength DIAL or simultaneous measurement of several substances by two wavelength DIAL. In this paper we present a description of the multiwavelength DIAL system, and report results of ozone concentration measurement in the lower troposphere by two identical DIAL pairs and analyze the various sources of measurement error.

II. Multiwavelength DIAL System

A schematic diagram of the multiwavelength DIAL system is given in Figure 1 and the specifications of the transmitter and receiver are shown in Table 1. The multiwavelength DIAL transmitter consists of two dye lasers (HOYA Continuum ND6000), each pumped by the second harmonic of two Nd:YAG lasers (HOYA Continuum Powerlite 8010) each operating at a repetition rate of 10Hz. The dye laser outputs can be doubled by second harmonic generators (SHG), so that the transmitter is tunable from the ultraviolet to the near infrared. Moreover, each dye laser is capable of emitting two wavelengths (λ_a, λ_b) on alternate pulses, each wavelength being tunable within the

Transmitter	
Pulse energy	>30mJ @300nm
Repetition rate	10Hz \times 2
Spectral width	<0.08cm ⁻¹
Stability	0.5pm/ ^o C/hr @300nm
Beam divergence	<0.5mrad (before beam expander) <0.1mrad (after beam expander)
Receiver	
Telescope type	Newtonian
Telescope diameter	500mm
Field of view	0.5~3.0mrad
Telescope reflectivity	75% @300nm
Filter transmission	48% @290nm
(for O ₃ measurement)	41% @285nm
Photomultiplier quantum efficiency	27% @300nm
Digitizer resolution	12bits
Digitizer rate	20MHz

Table 1: Specifications of the multiwavelength DIAL system.

oscillation range of the dye in use. This is accomplished by a rotating flat in the dye laser oscillator cavity which can direct the beam to one of two tuning mirrors corresponding to different wavelengths. The relative timing Δt of the two pump lasers is variable and can be set by a delay generator (Stanford Research Systems DG535) so that the transmitter can emit a sequence of four wavelengths consisting of a wavelength pair separated by Δt , followed 100ms later by a different wavelength pair, also separated by Δt , as shown in Figure 2 (Δt is set to 100 μ s in the figure). This four-wavelength sequence can be

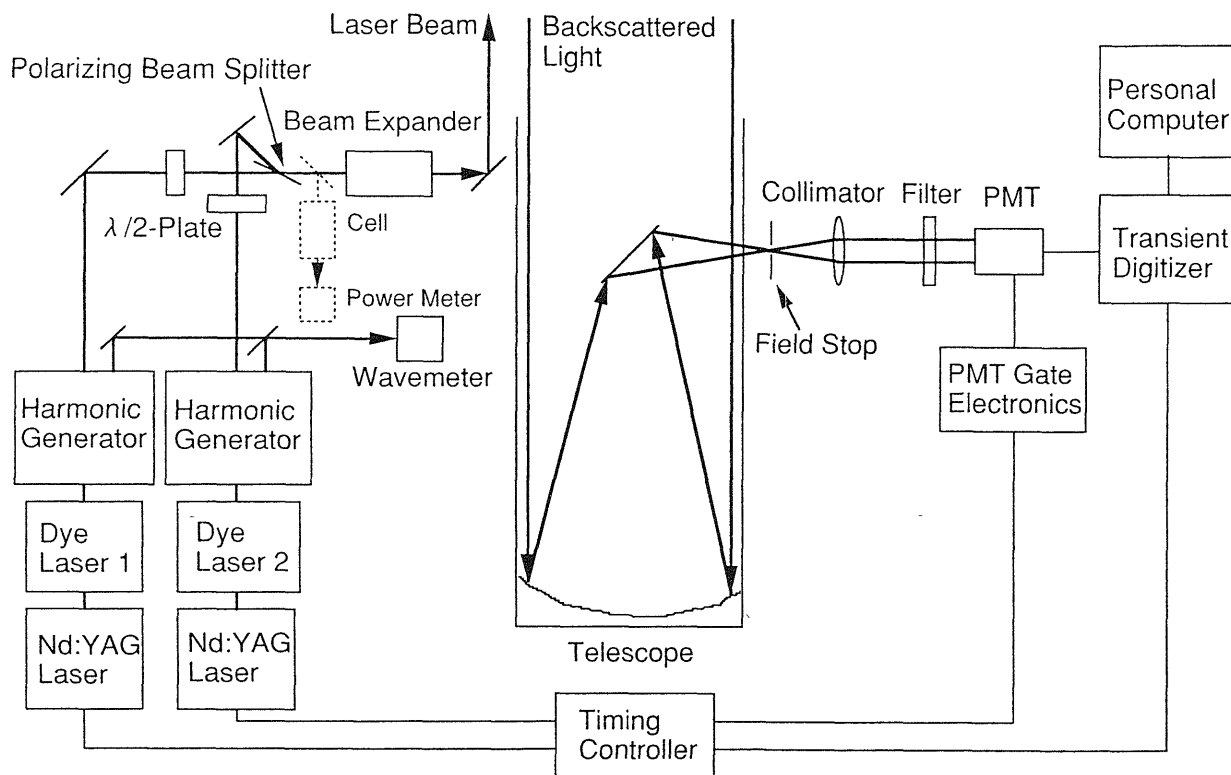


Figure 1: Schematic of multiwavelength DIAL system.

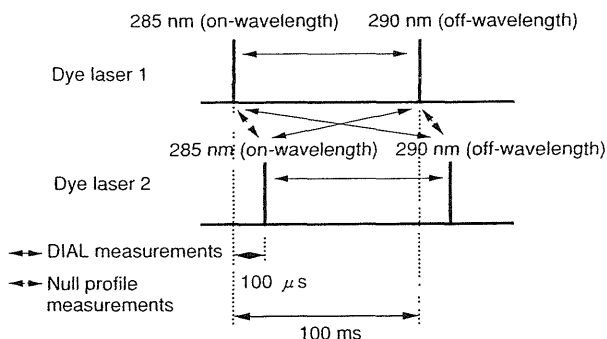


Figure 2: Timing of the two lasers.

used for multiwavelength DIAL measurement or two simultaneous DIAL measurements.

The SHG consists of two crystals each of which functions as the doubler for λ_a and λ_b , so that phase matching for frequency doubling at two wavelengths is accomplished without mechanical movement of the crystals. The two beams are combined using a polarizing beam splitter and the combined beam is expanded fivefold before being transmitted vertically into the atmosphere. The beam divergence is $<0.5\text{mrad}$ at the SHG output and $<0.1\text{mrad}$ after expansion. For ozone measurements, Rhodamine dye in methanol was used.

The receiver consists of a 50cm diameter Newtonian telescope which focuses backscattered radiation to a field stop whose field of view is variable between 0.5 and 3.0mrad. The light is then collimated, passes through an interference filter, and is converted to electrical signal by a photomultiplier tube (PMT, Hamamatsu 3896). For ozone measurement, an interference filter of center wavelength 290.5nm and bandwidth 19.0nm was used, and its transmission is 48% at 290nm and 41% at 285nm. The net telescope reflectivity is 75% and the PMT quantum efficiency is 27%, resulting in a total optical efficiency of 8~10% at the wavelengths used. The PMT signals are digitized by a 12bit, 20MHz digitizer (GaGe CS1012) corresponding to a range resolution of 7.5m.

III. Ozone Measurement

Two sets of experiments were performed. First, two simultaneous DIAL measurements using the same wavelength pairs were performed in order to check the effect due to laser beam alignment. The two dye lasers, labeled #1 and #2, were set to emit alternately at $(\lambda_{1a}, \lambda_{1b})$ and $(\lambda_{2a}, \lambda_{2b})$, respectively. The time delay between the pump lasers was set so that #2 was delayed by 100 μs with respect to #1. Therefore, the sequence of four wavelengths consisted of the

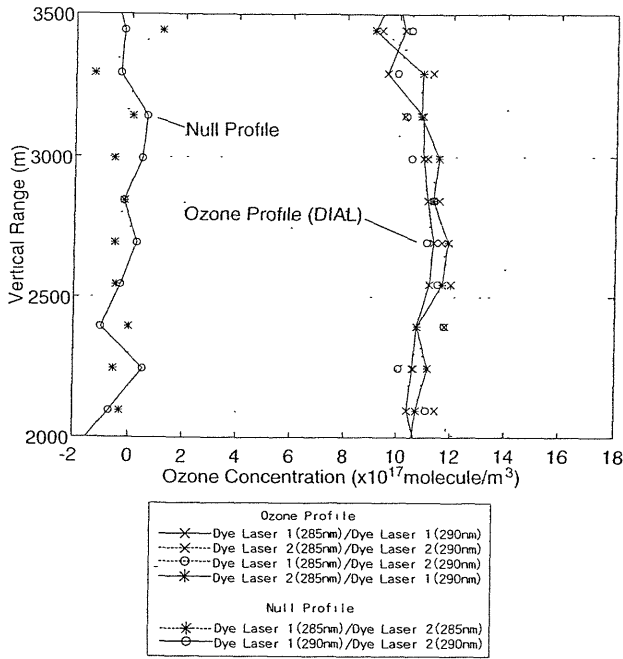


Figure 3: Vertical profiles of ozone concentration.

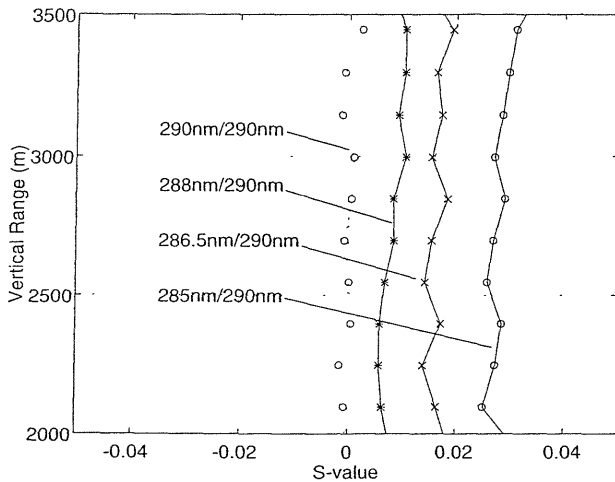


Figure 4: Vertical profiles of S-value for different wavelength pairs.

pair $(\lambda_{1a}, \lambda_{2a})$ separated by $100\mu s$, followed $100ms$ later by the pair $(\lambda_{1b}, \lambda_{2b})$, also separated by $100\mu s$. The wavelengths were set to $\lambda_{1a}=\lambda_{2a}=285.0nm$ and $\lambda_{2a}=\lambda_{2b}=290.0nm$, so that two null profile measurements $(\lambda_{1a}, \lambda_{2a})$, $(\lambda_{1b}, \lambda_{2b})$ and four DIAL measurements $(\lambda_{1a}, \lambda_{1b})$, $(\lambda_{1a}, \lambda_{2b})$, $(\lambda_{1b}, \lambda_{2a})$, $(\lambda_{2a}, \lambda_{2b})$, can be obtained simultaneously. The return signals were averaged over 5 minutes, corresponding to 1500 four-wavelength sequences or 6000 total shots.

To obtain the vertical concentration profiles, the obtained return signals were processed as follows: (1) the background level was subtracted, (2) all signals were averaged over 20 time bins corresponding to a

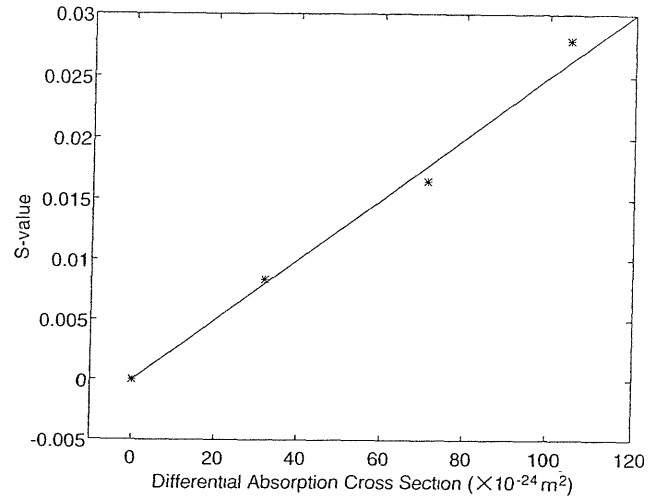


Figure 5: Plot of S-value vs. differential absorption cross section for different wavelength pairs.

range resolution of $\Delta R=150m$, (3) the S-value, given by

$$S_{ij} = \ln \left[\frac{P(R, \lambda_i)}{P(R + \Delta R, \lambda_i)} \frac{P(R + \Delta R, \lambda_j)}{P(R, \lambda_j)} \right] \quad (1)$$

was calculated for each of the six wavelength pairs (λ_i, λ_j) corresponding to the two null profiles are four DIAL profiles, and (4) the ozone concentration was calculated from

$$n = \frac{S_{ij}}{2\Delta R\Delta\sigma} \quad (2)$$

where $\Delta\sigma = 1.1 \times 10^{-24} m^2$ is the ozone differential absorption cross section for 285.0nm and 290.0nm. For the two null profiles, the same value of $\Delta\sigma$ was used for comparison with the DIAL profiles. In theory, if the beams profiles were identical and if the beams were precisely aligned, the S-value and concentration for the null profiles should be identically zero in the absence of statistical error. Therefore, the average deviation of the null profile from zero will provide a measure of how precisely the beams were aligned.

The obtained profiles for 2000~3500m altitude are shown in Figure 3. The average of the null profiles for this height interval is $5.6 \times 10^{15} molecule/m^3$, compared to typical DIAL values of $1.1 \times 10^{18} molecule/m^3$. In addition, the four DIAL pairs show the same result, indicating that all laser beams were precisely aligned, and systematic error caused by misalignment was low.

For the second set of DIAL experiments, λ_a was fixed at 290.0nm and λ_b was set to 290.0nm, 288nm, 286.5nm, 285.0nm, corresponding to different differential absorption cross sections. The measured S-values can be used to check the linearity of the receiver. The obtained vertical profiles of the S-value

Factors	Errors	
	$\times 10^{16}$ molecule/m ³	%
Inaccuracy of differential absorption cross section	2.2	2.0
Statistical error	5.1	4.6
Absorption by other molecules and particles	0.25	0.2
Null error (instrumental error)	0.56	0.5
Total	5.6	5.1

Table 2: Averaged errors of ozone concentration measurement for altitude 2000~3500 m. The percentage values are relative to the measured ozone concentration of 1.1×10^{18} molecule/m³, averaged for altitude 2000~3500m.

for 2000~3500m altitude are shown in Figure 4. The height-averaged S-value is plotted against differential absorption cross section in Figure 5, which verifies the linearity of the receiver. This shows that the same ozone concentration (8.3×10^{17} molecule/m³) was measured using different wavelength pairs.

IV. Error Analysis

The measurement errors are due to several factors: (1) inaccuracy of the differential absorption cross section data used in calculating the concentration, (2) statistical error, (3) effect due to other molecules and particles, and (4) instrumental error. These errors were evaluated for the profiles given in Figure 3. The inaccuracy of ozone absorption cross section was estimated from the difference in several published values and the wavelength stability of the dye lasers to be about 2%. The statistical error was estimated from the standard deviations of measured concentrations obtained for the four DIAL wavelength pairs, which was 5.1×10^{16} molecule/m³, or 4.6% relative to the average ozone concentration of 1.1×10^{18} molecule/m³. Since the beams were precisely aligned, the four measurements can be considered to be four independent, simultaneous measurements of the ozone concentration, so their deviation can be considered as being statistical in nature. For the effect due to other molecules and particles, the difference in Rayleigh and Mie scattering and extinction coefficients at the two wavelengths (285.0nm, 290.0nm) was considered, resulting in an equivalent ozone concentration of 2.5×10^{15} molecule/m³, or 0.2%. Interference due to other species such as SO₂ were considered insignificant. The instrumental error was estimated from the average deviation from zero for the two null profiles, which equalled 5.6×10^{15} molecule/m³, or 0.5%. The total error was obtained by adding the squares of these four errors and taking the square root, and found to be 5.1%. These values are summarized in Table 2.

V. Conclusion

In summary, a multiwavelength DIAL system for measurement of trace atmospheric substances was developed. The system is capable of emitting a sequence of four wavelengths, which can be used for high-resolution, multiwavelength DIAL measurement of a single species or simultaneous DIAL measurement of two or more species. Ozone measurement in the lower troposphere was performed using two identical DIAL pairs, from which two null profiles and four DIAL profiles were obtained. The null profiles showed that instrumental error due to beam misalignment was low (0.5%). The DIAL profiles showed an average ozone concentration of 1.1×10^{18} molecule/m³, and the total measurement error was about 5%. Multiwavelength measurement of ozone using different DIAL pairs (dual-DIAL measurement) is of further work.

References

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