signal intensity is plotted as a function of the mixing ratio with N_2 gas with a total pressure of 1 atm. The detection limit is estimated to be about 2 ppm for H_2.

In the case of CH_4, the PARS signal down to about 10 ppm level have also been detected by changing the Raman gas to CH_4. Further optimization to improve the sensitivity is now going on. Because the pump laser energy in our experiment is much higher than those by Siebert, *et al.*,[2] we can expect sensitive detection of CH_4 under 1 ppm level.

In summary, we proposed and demonstrated trace gas detection in the atmosphere, using a novel type of photoacoustic Raman spectroscopy with no tuning device. The proposed scheme can be applied to many other Raman-active gases, and suitable for long-term monitoring of the atmosphere because the system is simple and reliable. Sensitive detection of inflammable gas such as H_2 and CH_4 is also useful as a gas-leak monitoring system in the chemical plants and gas pipe lines.

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ThJ3 **1145**

Laser long-path absorption experiments using the Retroreflector in Space (RIS) on the ADEOS satellite

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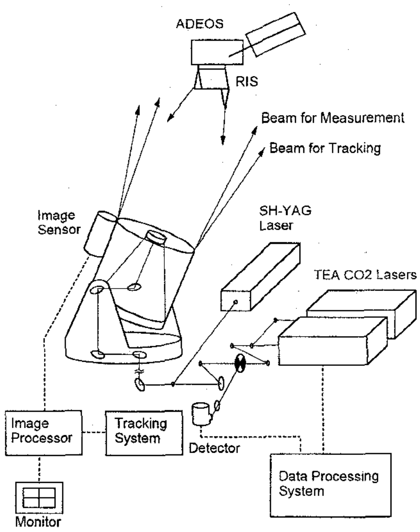
Earth-satellite-earth laser long-path absorption experiments have been carried out with the Retroreflector in Space (RIS) on the Advanced Earth Observing Satellite (ADEOS) which was successfully launched in August of last year by the National Space Development Agency of Japan (NASDA). The RIS is a 0.5 m diameter single-element hollow cube-corner retroreflector with a unique design having a curved surface for correcting velocity aberration caused by the satellite movement.[1]

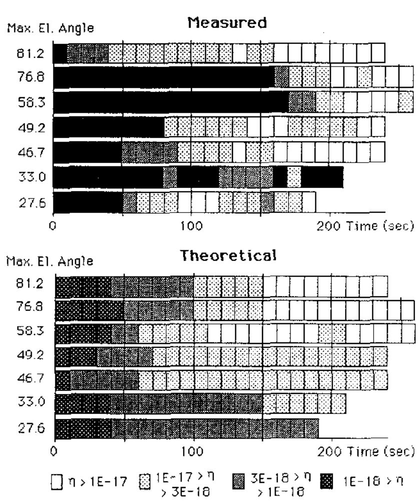
The ground system for the experiment consists of an optical satellite tracking system with 1.5 m diameter telescope and a laser transmitter/receiver system using two single-longitudinal-mode TEA CO_2 lasers and a second-harmonics Nd:YAG laser. Figure 1 shows a block diagram of the transmitter/receiver system. The YAG laser is used for active tracking using the image of reflection of the RIS. It is also used for laser ranging to the ADEOS. Spectroscopic measurement is carried out with the two TEA CO_2 lasers. One of the lasers is used for measuring absorption of atmospheric trace gases, and the other is used for recording a reference signal. A method using the Doppler shift of reflected beam caused by the satellite movement was developed for the RIS experiment.[2] In the spectrum measurement, pulse shapes of transmitted and received CO_2 laser pulses are recorded every shot. Repetition rate of the CO_2 lasers is 50 Hz, and time interval between pulses from two lasers is 200 μs. The pulse energy is approximately 100 mJ.

After the launch of the ADEOS, efficiency of the reflection of the RIS in orbit was checked first. Measured efficiency of the RIS at 532 nm is compared with the theory[3] in Fig. 2. They agreed well. It was also confirmed that intensity of infrared return at 10 μm agreed with the theory. An example of transmitted and received CO_2 laser pulse shapes is shown in Fig. 3.

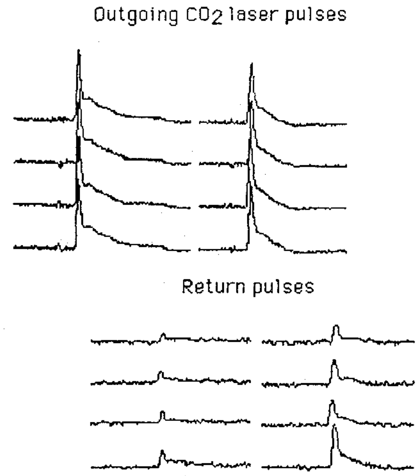
Figure 4 shows a preliminary atmospheric spectrum measured with the RIS by means of the Doppler shift method. Two CO_2 lasers were tuned to 9P(24) of $^{12}CO_2$ and 10R(24) of $^{13}CO_2$, respectively. Logarithm of the ratio of the signals is indicated as a function of time during the scan that corresponds to the wavelength of reflected laser beam. An absorption line of ozone is seen in the spectrum. Error in the measured spectrum is probably caused by the error in satellite tracking, change in transmitted laser beam pattern, and effects of atmospheric turbulence. Study is being carried out to optimize the optics to reduce the error in the spectrum measurement.



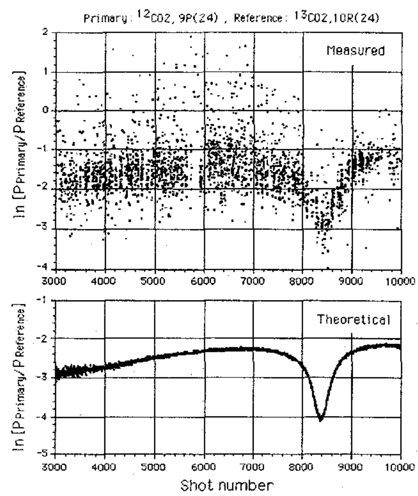
ThJ3 **Fig. 1.** A schematic diagram of the ground system.



ThJ3 **Fig. 2.** Measured efficiency of the RIS (a) compared with the theory (b).



ThJ3 **Fig. 3.** Transmitted and received CO_2 laser pulses.



ThJ3 **Fig. 4.** Absorption spectrum of ozone measured with the RIS.

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ThJ4 **1200**

Trace gas detection using mid-infrared laser difference-frequency generation

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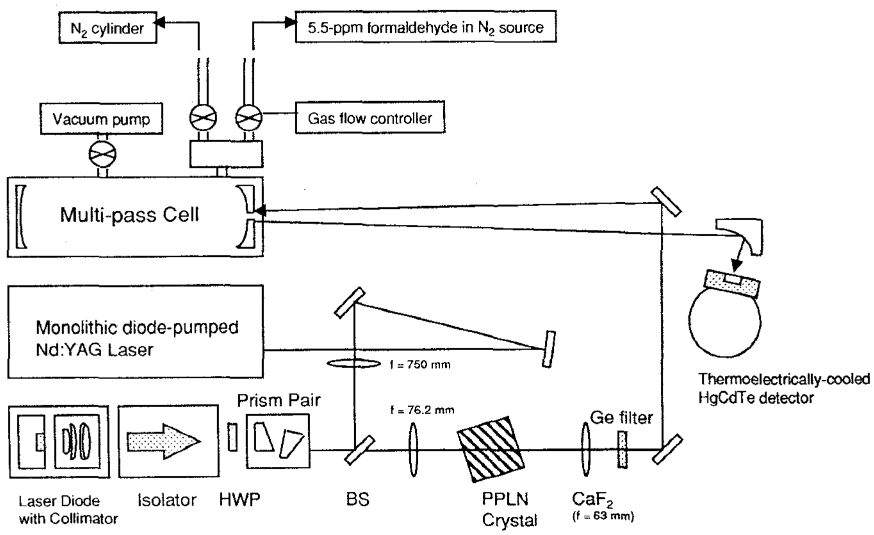
The detection and monitoring of trace and hazardous gases such as methane

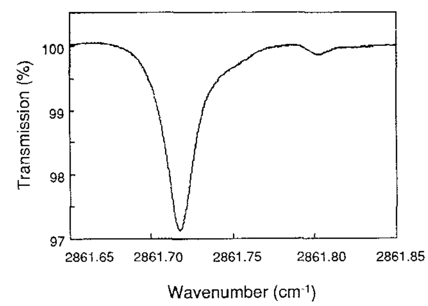


ThJ4 Fig. 1. Schematic diagram of the laser-based formaldehyde sensor. HWP: half wave plate, BS: beam splitter.

(CH_4), formaldehyde (H_2CO), and hydrogen chloride (HCl) is of considerable interest. Methane plays an important role in the greenhouse effect, while formaldehyde is a mediator in hydrocarbon oxidation in the troposphere. Both hydrogen chloride and formaldehyde are hazardous toxic gases that can be released in industrial processes. Methane and formaldehyde[1] absorption lines are available over a broad range of the mid-infrared region, while hydrogen chloride has strong and isolated lines around 3.5 μm. All three gases allow the use of a direct absorption method to measure atmospheric concentrations at the sub-ppm level in ambient air.

Recently, detection of methane and carbon monoxide (CO) by diode-pumped difference-frequency generation in periodically poled $LiNbO_3$ (PPLN) was demonstrated.[2,3] This method, in conjunction with a solitary or an external-cavity diode laser, can be applied to the detection of other gases by simultaneous adjustment of the diode laser wavelength and the PPLN domain grating period.

Here we have applied this method to the detection and measurement of formaldehyde. A schematic diagram of the sensor configured for the detection of formaldehyde is shown in Fig. 1. The sensor consists of two compact pump sources, a 100 mW single-frequency GaAlAs diode laser at 815 nm and a 700 mW single-frequency monolithic ring Nd:YAG laser whose output beams are overlapped in a 10 mm long PPLN crystal with a 21 μm domain grating period. The crystal can be rotated about the optic axis to form an angle of 21 degrees between the grating **k**-vector and the beam within the crystal, giving an effective grating period of 22.5 μm, as required for quasi-phase-matching. The DFG source can generate idler output powers of up to 4.7 μW at 3.5 μm. The idler wavelength is tunable over approximately 0.8 cm^{-1} by the laser diode current modulation. This tuning range is sufficient for monitoring a single absorption line of formaldehyde in air at atmospheric pressure. A gas mixing and flow controlling system is used to obtain gas mixtures at sub-ppm levels from a commercial 5.5 ppm formaldehyde-in-nitrogen mixture.



ThJ4 Fig. 2. A spectrum of 5.5-ppm formaldehyde at 2861.72 cm^{-1} in 50 Torr of nitrogen. A weak line at 2861.78 cm^{-1} assigned in reference 1 is also observed. The sampling time is ~60 seconds.

The idler beam collimated by the CaF_2 lens passes through a compact multi-pass cell for increased optical absorption. This cell has an effective path length of 18 m with an optical throughput of 35%. The idler beam emerging from the cell is collected by an off-axis parabolic mirror and focused onto a thermoelectrically-cooled HgCdTe detector. The detected signal is amplified by a dc-coupled preamplifier and processed with a laptop computer equipped with a PCMCIA 16-bit analog-to-digital converter.

A spectrum of a 5.5-ppm formaldehyde in 50 Torr of nitrogen at ~2861 cm^{-1} is depicted in Fig. 2. The line profile at 2861.72 cm^{-1} does not change when nitrogen is replaced with air, because no appreciable interference with other absorption lines is observed. The wavelength calibration for Fig. 2 was conveniently determined from methane spectra obtained prior to the H_2CO measurements. The minimum detectable formaldehyde concentration estimated from performance of the sensor in its present configuration is 6 ppb. It is limited by optical interference due to imperfect alignment and the preamplifier noise. Both can be reduced, making it possible to detect even lower concentrations. We also observed the spectrum of formaldehyde at atmospheric pressure. The sensor can also detect the fundamental absorption line of hydrogen chloride at 2843.6 cm^{-1}, if the diode wavelength is adjusted to 817 nm and the PPLN grating period to 22.6 μm.

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ThJ5 **1215**

Performance characterization of differential absorption lidar using radar perspectives

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The Differential Absorption Lidar (DIAL) in its several implementations has found applications in many aspects of optical remote sensing of the atmosphere, including pollution monitoring and environmental studies. Most standard formulations describing performance, however, do not allow ready evaluation of the interactions of lidar parameters, the ambient atmosphere, and the characteristics of the gas that is to be sensed. As a result, a preferred wavelength for detection of a candidate molecule, and the laser transmitter that might be most suitable for interrogation in the required atmosphere, are not always clear.

We have adopted the standard radar range equation as described by Skolnik[1] for meteorological echoes, and equivalently by Nathanson[2] to predict detection performance of a non-coherent DIAL that uses aerosol backscatter as a reference. This relationship has been used to predict the range at which NO_2 concentrations, with 82 ppm typical of that in the diluted exhaust of a large diesel engine,[3] could be detected in various conditions using a frequency-doubled Ti:sapphire DIAL system operating in the blue-visible with nominal parameters shown in Table 1. Results of the range calculations are shown in Fig. 1 for combinations of visibility and pulse energy, with 200 mJ the nominal value.

These results are quite optimistic, since they do not include the pseudo-noise effects induced by localized differences in differential backscatter and path absorption, nor are speckle and atmospheric turbulence explicitly considered. That is, the small difference signal from the candidate gas must be detected in the presence of a fluctuating clutter reference whose frequency spectrum also contains the desired signal. Fortunately, the transit time of small scale eddies at likely wind velocities place most of the clutter power at frequencies less than a few kHz.