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Possibility of hard-target lidar detection of a biogenic volatile organic compound, α -pinene gas, over forest areas

Yasunori Saito, Petter Weibring, Hans Edner, and Sune Svanberg

The absorption spectrum of α -pinene gas, a biogenic volatile organic compound, was directly measured with a pulsed mid-infrared laser. The maximum absorption wavelength was found to be $\sim 3.42 \mu\text{m}$, and an absorption cross section of $4.8 \times 10^{-23} \text{ m}^2 \text{ molec}^{-1}$ was obtained. A simple theoretical calculation with the measured spectral data showed that several hundreds of parts in 10^{12} (ppt) of α -pinene gas in forest-mountain areas over a range of $\sim 10 \text{ km}$ were detectable by a long-path-averaged hard-target absorption lidar. Requirements for system development were also discussed. © 2001 Optical Society of America
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Biogenic volatile organic compounds (BVOCs) emitted from vegetation¹ play an important role in the formation and removal of atmospheric molecules. The gaseous BVOCs are so reactive that complex interactions between BVOCs and other natural-artificial compounds generate new compounds. For example, ozone is formed through a reaction of NO_x and BVOCs within the planetary boundary layer.² BVOCs have a significant effect on the balance of the atmosphere directly and indirectly. Remote-sensing techniques can be effective aids for understanding the dynamics of BVOCs in an area of forests and mountains that is spread over several kilometers. Also, as the BVOCs are quite active and short lived, it is strongly recommended that experiments such as concentration measurements be done *in situ* and under natural conditions rather than being done in laboratories with samples.

In this short paper we discuss the possibility of lidar detection of gaseous α -pinene by showing calculation results from use of measured absorption data of α -pinene, which is one of the major BVOCs in northern

European forests.^{3,4} For remote molecule detection it is anticipated that an absorption-type lidar and a Fourier-transform-type lidar⁵ will meet the requirements. In particular, a differential-absorption lidar based on a solid-state optical parametric oscillator technique⁶⁻⁸ would be more practical if the target molecule (α -pinene in this case) were selected and measurement of an area of several kilometers within 1 h were required, where the concentration of α -pinene was less than parts in 10^9 , on variation of the daily changes in vegetation.^{3,4,9,10} Such measurements of the α -pinene concentration would be used for making a more detailed and realistic model of photochemistry that can express phenomena that occur between the biosphere and the atmosphere. Also, it could be used as an indicator of changes in vegetation and an estimation of biomass resources for planning and management of a forestry environment.

An optical parametric oscillator system incorporating a Nd:YAG laser was developed for absorption experiments.¹¹ Mid-infrared laser output was generated from a mixture of the fundamental Nd:YAG output of $1.064 \mu\text{m}$ and tunable near-infrared optical parametric oscillation radiation near $1.5 \mu\text{m}$ in a LiNbO_3 crystal. We tuned the wavelength in the mid-infrared region near $3.4 \mu\text{m}$ by scanning the near-infrared wavelength automatically with a program under the control of a personal computer. The energy of the mid-infrared laser pulses was $\sim 0.5 \text{ mJ}$, the pulse width was 3 ns, and the repetition rate was 20 Hz. The laser output was split into two beams. One beam was passed through an absorption cell filled with α -pinene, and the other one was used as a

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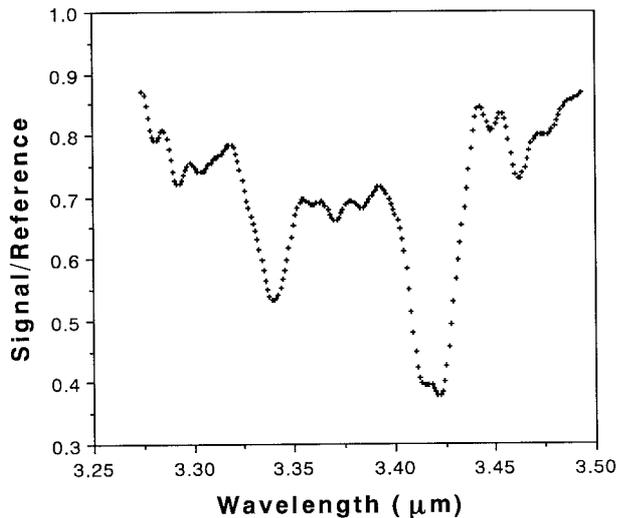


Fig. 1. Transmittance spectrum of α -pinene gas.

reference. The energy of the laser beam was adjusted by neutral-density filters to set the signal beam and the reference beam to have similar energy at the detectors. The signal and the reference beams were detected with two InGaAs detectors. A set of Ge filters was used to eliminate the residual near-infrared beam and the Nd:YAG laser with the addition of a LiNbO₃ crystal, and a CaF₂ lens was used to focus the laser beams onto the detectors. The absorption cell was 0.94 m long, with an inside diameter of 40 mm. Each end of the cell had a sapphire window. A bottle of liquid α -pinene (Merck-Schuchardt, Munich) was opened in a plastic bag that had been filled with nitrogen gas at 1×10^5 Pa (1 atm). The vapor pressure of α -pinene was 5 hPa at a temperature of 25° and a pressure of 1×10^5 Pa. After the absorption cell was evacuated, a measured amount of the α -pinene and nitrogen mixture from the bag was introduced into the cell, and the gas pressure inside the cell was monitored with a vacuum pressure gauge with a resolution of 0.1 Torr on its scale. Finally, nitrogen gas was added to the cell for a total pressure of 1×10^5 Pa.

Figure 1 shows a measured transmission spectrum (the signal divided by the reference) of α -pinene near 3.3–3.5 μm . In this experiment the pressure of α -pinene was adjusted to yield as much as 50% absorption in the wavelength range studied. The data points were averages of 64 pulses and were separated by 1.1 nm. The strongest absorption peak appeared near 3.422 μm . The dependence of transmittance on pressure is illustrated in Fig. 2, for which the wavelength was fixed at the strongest absorption peak. The calculated absorption coefficient was 4.8×10^{-23} m² molec⁻¹. Although there was a small drift in the vacuum levels, a partial α -pinene gas pressure of as little as 3.3 Pa could be seen.

We now discuss hard-target lidar detection of α -pinene gas over forest–mountain areas, based on the data obtained from the absorption experiments. Following Killinger and Menyuk,¹² the minimum de-

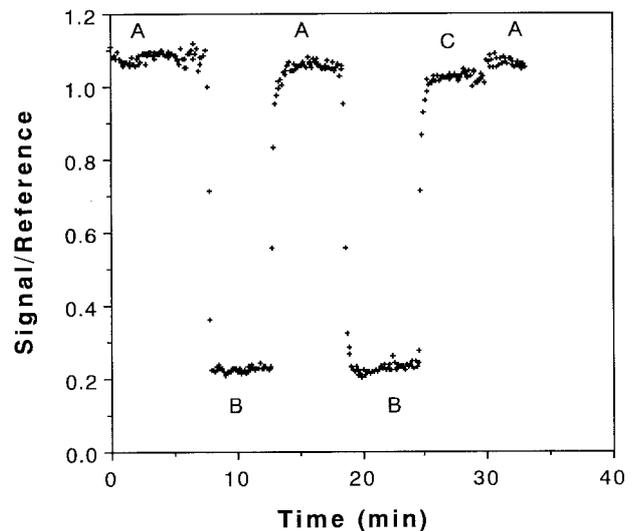


Fig. 2. Dependence of the transmittance on pressure in α -pinene gas at the maximum absorption wavelength. A, vacuum. Total cell pressure of 1.0×10^5 Pa (1 atm) containing B, 1.3×10^2 Pa of α -pinene and C, 3.3 Pa of α -pinene.

tectable concentration (N_{\min}) for a mid-infrared long-path-averaged differential absorption lidar (DIAL) is associated either with (1) detector-noise limitation or (2) changes in atmospheric conditions, not including absorbing molecules. In case (1), N_{\min} for the DIAL measurement is approximately determined by the detector's noise-equivalent power (NEP) as

$$N_{\min} \cong \frac{(\text{NEP})\pi R}{2K\rho AP_t\sigma \exp(-2\beta'R)},$$

where R is the range from the system to a hard target, K is the overall optical efficiency of the system, ρ is the reflectivity of the target, A is the area of the receiving telescope, P_t is the transmitted power, σ is the absorption cross section of the absorbing gas, and β' is the background atmospheric-extinction coefficient, not including the absorbing gas. For the equation it is assumed that $2\sigma N_{\min}R \ll 1$. In case (2) the uncertainty in the measurement is determined by the minimum fractional change in the return signal, $\Delta P/P'$, as

$$N_{\min} \cong \frac{(\Delta P/P')}{2\sigma R},$$

where $\Delta P = P' - P$; P' and P are the received instantaneous powers of the DIAL measurement at the nonabsorption wavelength and the absorption wavelength, respectively. The assumption is made that $2\sigma N_{\min}R \ll 1$.

The estimated N_{\min} of α -pinene is shown in Fig. 3 as a function of range for two cases. Parameters for the calculation were NEP of 7×10^{-9} W, $K = 0.6$, $\rho = 0.1$ for a topographic target and $\rho = 1.0$ for a retro-reflector, $A = 0.2$ m², $P_t = 1 \times 10^6$ W (5-mJ pulse energy; 5-ns pulse width), $\sigma = 4.8 \times 10^{-23}$ m², and $\beta = 2.2 \times 10^{-4}$ m⁻¹ (clear sky).¹³ It can be seen that

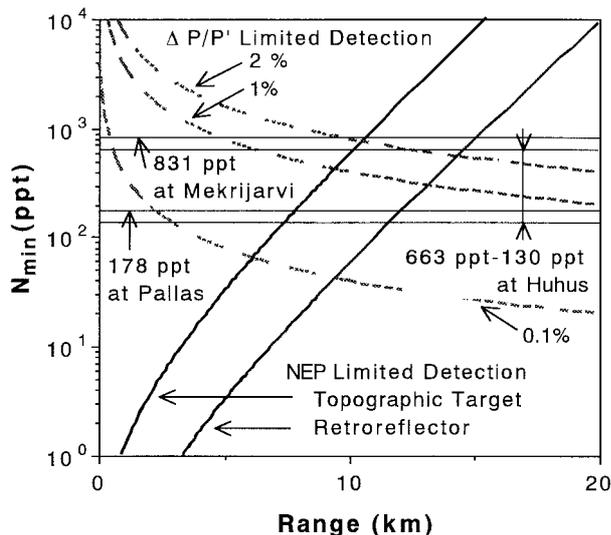


Fig. 3. Minimum detectable concentration of long-path-averaged lidar detection of α -pinene gas as a function of range estimated by NEP limited detection and differential-return $\Delta P/P'$ limited detection.

the fractional change in the return signal $\Delta P/P'$ limits the sensitivity of the measurement at short range; the NEP limitation, however, increases with the range. The estimated values of N_{\min} were compared with the actual value. The concentration of α -pinene strongly depends on meteorological conditions and sites. For example, it was an average of 831 parts in 10^{12} (ppt) at Mekrijarvi and 178 ppt at Pallas,³ Hakola *et al.* reported a concentration that varied up to 768 ppt at Ilomants,⁹ and Rinne *et al.* reported a variation from 663 to 130 ppt in 8 h at Huhus.¹⁰ These values are taken from the references cited here. All the sites are located in the boreal zone of Finland, and sample air was analyzed with a gas chromatograph.⁴ For a topographic target, the minimum fractional change of $\Delta P/P'$ should be less than 2% to correspond to the change in the α -pinene concentration, depending on the test site. If $\Delta P/P'$ is 1%, N_{\min} is less than 831 ppt within a range of 5–10 km. If a retroreflector could be used, the detectable range for 1% ($\Delta P/P'$) would expand to 13 km. Such a range can cover an entire mountain area. At Huhus, a part of the variation of the concentration under this condition of 1% ($\Delta P/P'$) can be observed. In the figure, 0.1% ($\Delta P/P'$), which was the digitization error level of the analog-to-digital converter obtained by the number of pulses integrated into a laboratory experiment,¹² was included as an assumed detection limit. In an outdoors experiment, in which the pointing stability of the laser beam is influenced by atmospheric turbulence,¹⁴ it is important to consider the value of $\Delta P/P'$. Such good pointing stability will be achieved by introduction of an active tracking technique, as was successfully done in the Earth–satellite–Earth long-path absorption experiment with a retroreflector.¹⁵

We experimentally investigated the absorption

characteristics of α -pinene gas by using laser absorption techniques. Comparing the estimated results and the expected actual concentration of α -pinene showed the possibility of lidar detection of α -pinene over forest–mountain areas. A mid-infrared lidar system that can scan over a specific absorption spectral region can be used for resolving spectral interference effects between the molecules of interest and others by a multivariate statistical technique.¹¹

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