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Possibility of hard-target lidar detection of a biogenic volatile organic compound, \(\alpha\)-pinene gas, over forest areas

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The absorption spectrum of \(\alpha\)-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 \(\alpha\)-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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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 LiNbO$_3$ crystal, and a CaF$_2$ 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 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$^{-23}$ m$^2$ 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 detectable concentration ($N_{\text{min}}$) 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), $N_{\text{min}}$ for the DIAL measurement is approximately determined by the detector’s noise-equivalent power (NEP) as

$$N_{\text{min}} \approx \frac{\text{NEP} \pi R}{2K_\rho AP_\sigma \exp(-2\beta' R)},$$

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

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

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 $2\sigma N_{\text{min}} R \ll 1$.

The estimated $N_{\text{min}}$ of α-pinene is shown in Fig. 3 as a function of range for two cases. Parameters for the calculation were NEP of $7 \times 10^{-9}$ W, $K = 0.6$, $\rho = 0.1$ for a topographic target and $\rho = 1.0$ for a retro-reflector, $A = 0.2$ m$^2$, $P_\sigma = 1 \times 10^6$ W (5-mJ pulse energy; 5-ns pulse width), $\sigma = 4.8 \times 10^{-23}$ m$^2$, and $\beta = 2.2 \times 10^{-4}$ m$^{-1}$ (clear sky).$^{13}$ It can be seen that
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 \( N_{\text{min}} \) were compared with the actual value. The concentration of \( \alpha \)-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. Hakola et al. reported a concentration that varied up to 768 ppt at Iломants, and Rinne et al. reported a variation from 663 to 130 ppt in 8 h at Huhus. 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. For a topographic target, the minimum fractional change of \( \Delta P/P \) should be less than 2\% to correspond to the change in the \( \alpha \)-pinene concentration, depending on the test site. If \( \Delta P/P \) is 1\%, \( N_{\text{min}} \) 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, 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, 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.

We experimentally investigated the absorption characteristics of \( \alpha \)-pinene gas by using laser absorption techniques. Comparing the estimated results and the expected actual concentration of \( \alpha \)-pinene showed the possibility of lidar detection of \( \alpha \)-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.

**References**