Remote \( \text{SO}_2 \) measurements at \( 4 \, \mu \text{m} \) with a continuously tunable source

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Remote atmospheric measurements of \( \text{SO}_2 \) were performed using a differential absorption lidar with a continuously tunable \( \text{LiNbO}_3 \) parametric oscillator and amplifier source in the \( 4.0-\mu \text{m} \) region. A comparison of injected gas concentration in a remotely located sample chamber with the lidar measurements through the chamber showed measurement agreement within experimental error and a detection sensitivity for \( \text{SO}_2 \) of 0.9 part in \( 10^6 \) km.

This Letter presents results of remote atmospheric lidar measurements of \( \text{SO}_2 \) using a continuously tunable parametric oscillator and amplifier source in the \( 4.0-\mu \text{m} \) region. A sample chamber in the optical path of the lidar simulated a source of \( \text{SO}_2 \), allowing a direct comparison of lidar measurements with \textit{in situ} gas-concentration values.

In the present measurements we obtained a sensitivity of 0.9 part in \( 10^6 \) km with a transmitted pulse energy of 0.6 mJ at 10 pulses per second for 500 averaged pulses. Continuous tunability over the 1.4- to 4.0- \( \mu \text{m} \) range offers flexibility in selecting the optimum infrared spectral region and wavelength for pollutant measurements. This broad tuning range includes absorption bands of many pollutants, such as \( \text{CO}, \text{CO}_2, \text{CH}_4, \) and other hydrocarbons and \( \text{N}_2\text{O} \) in addition to \( \text{SO}_2 \). The details and background for differential-absorption and Raman-lidar techniques for remote monitoring are covered in review articles.1–3 The original measurement of \( \text{SO}_2 \) was made by Inaba,1 with several subsequent measurements in the ultraviolet.4,5

Because of the importance of \( \text{SO}_2 \) emission from stationary sources, especially from combustion of coal or fuel oil, a real-time remote measurement of \( \text{SO}_2 \) with infrared lidar is an important measurement capability.6 The full daylight operation with total eye safety is an advantage over the ultraviolet lidar system previously described by Grant et al.4 The capability of providing more positive spectral identification of a gaseous constituent with the continuous tuning is an advantage over discretely tuned infrared laser measurement.7 For example, numerous hydrocarbons with CH stretch resonances in the 3.3- \( \mu \text{m} \) region may be identified by scanning appropriate parts of their spectra. The continuous tunability of the present system offers the possibility of optimizing the measurement strategy for a given pollutant molecule by adjusting the absorption cross section and minimizing interferences.

The lidar is a monostatic system that uses topographic targets as retroreflectors. The beam is transmitted coaxially with a 40-cm-diameter Newtonian receiver telescope. A liquid-nitrogen-cooled InSb detector 1 mm in diameter, with a detectivity of \( 2 \times 10^{11} \) cm Hz\(^{1/2}\) W\(^{1/2}\), is located at the focal point. Atmospheric probing over a full hemisphere is possible with the fully steerable altazimuth telescope mount. The tunable source can be tuned on and off a molecular resonance for a differential absorption measurement.

Figure 1 shows a schematic diagram of the Nd:YAG-pumped \( \text{LiNbO}_3 \) parametric oscillator and amplifier that provides the pulsed infrared radiation over the tuning range of 1.4–4.0 \( \mu \text{m} \).8 The pump source is a 6.35-mm-diameter Nd:YAG unstable-resonator oscillator. Half of the 250-mJ output energy pumps the \( \text{LiNbO}_3 \) angle-phase-matched parametric oscillator through a Faraday rotator isolator. The remaining 125-mJ output energy is amplified in a 9.35-mm-diameter YAG amplifier and pumps a second 6-cm-long \( \text{LiNbO}_3 \) crystal parametric amplifier. The parametric oscillator is a singly resonant oscillator type with a double-pass pump wave. The parametric signal wave is resonated in a 13-cm-length cavity consisting of a 600-line/mm grating for wavelength tuning control and

![Fig. 1. Schematic diagram of the tunable source for the infrared lidar measurement.](image-url)
The sample chamber simulating the SO$_2$ source was located 120 m from the roof-mounted telescope. Oak trees, located an additional 75 m beyond the sample chamber, served as a topographic target. The chamber, borrowed from SRI International, has a path length of 2.44 m along the lidar direction, with low-density polyethylene windows mounted at a 15° angle to minimize direct backreflections into the receiver. Measurements were made by adding a known concentration of SO$_2$ to the chamber and measuring the concentration by scanning the lidar several times over the frequency range from 2945 to 2513 cm$^{-1}$, using the parametric idler wave with a 4-cm$^{-1}$ bandwidth. For normalization the chamber was purged of SO$_2$, and scanned providing the 100% reference values shown in Fig. 2. The error bars illustrating standard-deviation values were averages over three or four frequency scans and represent the overall response of the SO$_2$ spectrum and the system linewidth at each frequency point.

Figure 3 shows the excellent agreement between the sample-chamber gas concentration values and the lidar measurements over a large concentration range. The concentration values were derived by averaging frequency points over a region equivalent to the source linewidths (darkened regions in Fig. 2), on both a normalized on-resonance and off-resonance region. A system sensitivity may be derived by extrapolating to a unity signal-to-noise ratio. For the SO$_2$ measurements the value was 0.9 part in 10$^6$ km, as indicated earlier. The linewidth value of 3.5 cm$^{-1}$ for this measurement is quite appropriate since the SO$_2$-band linewidth is pressure broadened by the other atmospheric constituents.

Recent advances in parametric oscillator performance show that a substantial increase in output energy should

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\ln \left( \frac{P_{\text{off}}}{P_{\text{on}}} \right) = 2 \int_{0}^{L} P_{\text{SO}_2}(\alpha_{\text{on}} - \alpha_{\text{off}}) \, dr,
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where $P_{\text{off}}$ and $P_{\text{on}}$ represent the return power received off and on the molecular resonance, respectively, $L$ is the optical path length in the chamber, $P_{\text{SO}_2}$ is the partial pressure of SO$_2$ in the chamber, and $\alpha_{\text{on}} - \alpha_{\text{off}}$ is the on-resonance to off-resonance absorption coefficient. The absorption coefficient difference was obtained by a White-cell scan of SO$_2$ at 1-Torr partial pressure over a length of 675 cm. The measured value of $\alpha_{\text{on}} - \alpha_{\text{off}} = 0.363$ (atm cm)$^{-1}$ compares quite favorably with a value of 0.372 (atm cm)$^{-1}$ obtained in the same wavelength region by averaging the data of Pine and Moulton in a similar procedure used for the sample chamber data.
be possible. A transmitted energy of 15 mJ per pulse at 4.0 μm would improve the sensitivity of the SO₂ measurement to 30 parts in 10⁹ km for 500 averaged pulses, which is more than adequate sensitivity for source monitoring.

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