APPENDIX B:**MANUSCRIPT FOR THE OPTICAL REMOTE SENSING
IN THE ATMOSPHERE TOPICAL MEETING OF THE
AMERICAN CHEMICAL SOCIETY**

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Diode Laser-based Sensor System for Long-Path Absorption Measurements of Atmospheric Concentration and Near-IR Molecular Spectral Parameters.

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Line-locked near-IR diode lasers and a simple retroreflector/telescope system were used for remote sensing of atmospheric constituents over long atmospheric paths. Figure 1 shows the experimental configuration used in preliminary measurements of atmospheric water vapor and oxygen with AlGaAs diode lasers. A prototype field sensor system currently under development shares the same basic configuration but incorporates interchangeable AlGaAs and InGaAsP diode-laser modules for monitoring a variety of atmospheric gases.

Fig. 1

Fig.2

Fig.3

Absorption is detected via wavelength modulation and second harmonic detection. Figure 2 illustrates the variation of the second harmonic signal with the amplitude of modulation of the laser frequency about the line center. The data are for an atmospheric $^{16}\text{O}_2$ line at 13118.0 cm^{-1} . Theoretical fits of such spectra yield the line strength and pressure broadening parameters required to quantify the molecular absorption.

Figure 3 illustrates the quantification of $^{16}\text{O}^{18}\text{O}$ column density over distances ranging from 12 to 220 meters. Each data point corresponds to a measurement over a distinct atmospheric path defined by the stationary transceiver system and a movable retroreflector. Absorption of the P_9Q_8 line at 13097.3 cm^{-1} was quantified by the ratio of the second and first harmonic signals I_{2f} / I_{1f} at a fixed modulation amplitude. The I_{1f} signal at the line center is directly proportional to the transmitted intensity and independent of molecular absorption. The I_{2f} signal had a constant offset due to curvature in the diode-laser current-intensity relationship, and a component proportional to the absorption. Figure 3 shows the predicted dependence of the signal on path length based on literature values for spectral parameters and independent measurements of the temperature, laser modulation amplitude, and laser harmonic signals.

Fig. 4

Figure 4 shows simultaneous measurements of $^{16}\text{O}_2$ and H_2O over a common indoor path using two diode lasers. The absorbances at the line center were 0.6 and 0.08 respectively. Both species show diurnal and short-term variations in concentration. The range of oxygen absorption signals is consistent with the number densities associated with the 15° - 23° temperature range observed during the two day test. The gross trends in the water column density were confirmed by periodic wet/dry bulb hygrometer readings.

Figure 5 shows 24 hour column density measurements of the weak $^{16}\text{O}^{18}\text{O}$ P_9Q_8 line absorption over short path lengths of 1.7 and 11.9 m. The absorbance at the line center was 1.2×10^{-4} for the lower trace and 9×10^{-4} for the upper trace. Data points were collected at 5 minute intervals with a 1 second time constant. The long term drift visible in the lower trace was due to electronics baseline drift and had a standard deviation equivalent to an absorption of 1×10^{-5} . The standard deviation on a 10 minute time scale was a factor

Fig. 5

of ten lower. The upper trace shows additional variation due to changes in oxygen density with temperature. The solid lines represent the range of column densities associated with the measured temperature variation over the 24 hour period.

In conclusion, these measurements of water and oxygen lines demonstrate accurate and sensitive remote monitoring of concentrations and spectral properties of atmospheric constituents. This technique can be applied to a large number of atmospheric trace species absorbing in the near-IR. Spectral Sciences is currently developing a prototype instrument for automated, unattended operation in field studies of atmospheric composition. Progress toward that end and the application of the technique with InGaAsP diode lasers will also be discussed.

N. Goldstein, S. Adler-Golden, J. Lee, and F. Bien, "Measurements of Molecular Concentrations and Line Parameters using Line-locked Second Harmonic Spectroscopy with AlGaAs Diode Lasers," *Appl. Opt.*, **31**, 3409 (1992)