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ABSTRACT

A new optical tool, which will help petrochemical refinery and chemical processing plant personnel locate the source of a toxic or hazardous gas leak while remaining outside the perimeter of the processing area, has been demonstrated. This sensor is capable of locating leaks from a distance of about 20 m, with a response time of less than 1 second. By enhancing the speed with which leaks can be located, the risk of an incipient failure becoming a catastrophic failure is reduced significantly. The tool is based on Tunable Diode Laser Absorption Spectroscopy (TDLAS). TDLAS is currently being utilized in these industrial environments in permanently-installed open-path configurations to sense releases of selected gases as they exit the processing area. The sensor described here combines a lightweight, portable optical transceiver with battery-operated electronics in a single hand-held package. By standing in a safe area and “shining” the eye-safe laser beam emanating from this device onto suspected leak sources, operators may rapidly isolate the source while minimizing their potential exposure to the hazard. The sensor can be configured to sense leaks resulting in path-integrated concentrations of, for example, 2 ppm-m of hydrogen fluoride, 200 ppm-m of hydrogen sulfide, or 10 ppm-m of methane.

Keywords: TDLAS, spectroscopy, lasers, leak detection, methane, gas, sensors

1. INTRODUCTION

This paper describes a new optical tool which, when fully developed, will facilitate detection and location of toxic or hazardous gas leaks in petrochemical refineries, chemical processing plants, natural gas production facilities, and natural gas distribution pipelines. The tool is based on Tunable Diode Laser Absorption Spectroscopy (TDLAS), a sensing technology that is rapidly gaining acceptance in industrial environments for detecting releases of selected gases. Illustrated conceptually by Figure 1, when fully engineered the tool is expected to resemble a large flashlight. It illuminates a distant surface with laser light and measures the amount of target gas along the line of sight transited by the laser beam.

Unlike other types of portable gas detectors, this laser-based device does not need to be immersed within the gas leak. This is of value in, for example, chemical and petrochemical settings that operate tanks and pipelines containing hydrogen fluoride, hydrogen sulfide, ammonia, or methane. Although these facilities are operated with the strictest of safety precautions, equipment or human failure occasionally leads to a gas phase chemical release. Often, the leak sources such as flanges, valves, pump seals, and pipe pinholes are difficult to locate or access. The plant operator may receive an indication from an area sensor that a leak has occurred, but the sensor cannot pinpoint the source of the leak. During any such event, plant operators endeavor to first protect all personnel and then, as rapidly as possible, identify the source of the release and shut it off or instigate mitigation procedures, thereby limiting damage to the environment and surrounding communities. The envisioned laser-based sensor would allow plant personnel to isolate the source of a leak while remaining outside the perimeter of the processing area where the hazardous gas is highly concentrated. Thus, the risk to plant personnel is reduced, and by enhancing the speed with which leaks can be located, the risk of an incipient failure becoming a catastrophic failure is reduced significantly.

The work described below demonstrated experimentally that a portable TDLAS transceiver, sensing laser light that is transmitted through a gas cloud and scattered back to the transceiver from topographic surfaces beyond the gas cloud, can achieve a useful sensitivity to leaks of specific gases from a reasonable distance. The device was shown to sense specific target gases with a path-integrated concentration corresponding to a laser absorbance of about 8×10^{-5} , by detecting from a distance of about 20 m only the laser radiation scattered from surfaces located behind the leak source without a dedicated

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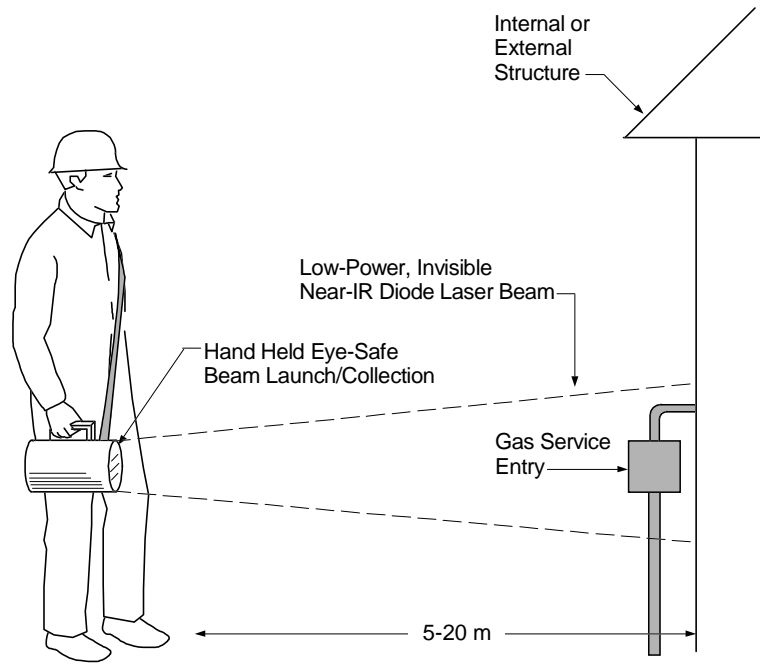


Figure 1. Illustration of portable TDLAS leak finding system.

retroreflector. This corresponds to an ability to locate toxic or hazardous gas leaks resulting in path-integrated concentrations of, for example, 1.6 ppm-m of hydrogen fluoride, 8 ppm-m of methane, or 160 ppm-m of hydrogen sulfide.

2. BACKGROUND

2.1. Tunable diode laser absorption spectroscopy

Tunable diode laser absorption spectroscopy (TDLAS) is a maturing technology that is enabling sensing trace concentrations of a number of critical gases in a broad array of applications.¹⁻⁵ During the past decade, the technology has emerged from the laboratory to become a reliable, robust, and commercially available means for continuously measuring and, in some cases, controlling extremely small concentrations of selected trace gas species.⁶⁻¹⁰

TDLAS instruments rely on well-known spectroscopic principles and sensitive detection techniques, coupled with advanced tunable diode lasers and optical fibers developed by the telecommunications industry.¹¹⁻¹⁴ The principles are straightforward: Gas molecules absorb energy at specific wavelengths in the electromagnetic spectrum. At wavelengths slightly different than these absorption lines, there is essentially no absorption. By: (1) transmitting a beam of light through a gas mixture sample containing a (usually trace) quantity of the target gas, (2) tuning the beam's wavelength to one of the target gas's absorption lines, and (3) accurately measuring the absorption of that beam, one can deduce the concentration of target gas molecules integrated over the beam's path length. This measurement is usually expressed in units of ppm-m.

The laser's fast tuning capability is exploited to rapidly and repeatedly scan the wavelength across the selected gas absorption line. While this scanning occurs, the fraction of emitted laser power that is transmitted through the gas mixture is monitored with a photodetector. The power transmitted across the measurement path, and thus the photocurrent generated by that path's detector, varies according to Beer's law

$$I_m(\nu) = I_{m0}(\nu)e^{-\alpha(\nu)}, \quad (1)$$

where $I_{m0}(\nu)$ = power transmitted as a function of wavenumber (the inverse of wavelength), $I_m(\nu)$ = power received as a function of wavenumber, and $\alpha(\nu)$ = path-integrated absorbance as a function of wavenumber. At atmospheric pressure, the spectral absorption feature shape is described by a Lorentzian function,

$$\alpha(\nu) = (SN_a L / \pi g) (P_g / P_a) [(1 / \{(v - \nu_0) / g P_a\}^2) + 1] \quad (2)$$

where

- S = spectral line strength (cm⁻¹/molecule-cm⁻²)
- N_a = 2.48 x 10¹⁹ molecules/cm³-atm
- g = linewidth at one atmosphere (cm⁻¹/atm)
- ν = wavenumber (cm⁻¹)
- P_a = total pressure of gas sample (atm)
- P_g = partial pressure of target gas (atm).
- L = length of optical path through the gas sample.

This procedure yields a precise and highly sensitive measure of the target gas concentration along the path transited by the laser beam. Thus, TDLAS instruments offer a combination of high sensitivity to trace concentrations of many gases, freedom from cross-species and external interference, and fast response. Examples of gases that can be sensed with TDLAS are listed in Table 1.

Table 1. Detection Limits (in ppm-meters) for Gases Measured with Near-IR TDLAS, Assuming Capability to Measure Absorbance of 1 Part in 10⁵ (i.e., a Noise Equivalent Spectral Absorbance or NESA of 10⁻⁵)

HF	0.2	HCN	1.0
H ₂ S	20.0	CO	40.0
NH ₃	5.0	CO ₂	40.0
H ₂ O	1.0	NO	30.0
CH ₄	1.0	NO ₂	0.2
HCl	0.15	O ₂	50.0

3. EXPERIMENTAL DESIGN

A breadboard prototype of the hand-held TDLAS sensor was built to experimentally evaluate key performance parameters. The sensor was built of three primary components: 1) a laser source and control electronics; 2) a transceiver; and 3) receiver electronics. The laser source and control electronics, as well as the receiver electronics, were assembled primarily from commercially-available laboratory equipment.

The basic optical configuration of the transceiver combines a laser collimating and transmitting section with a laser receiver and focusing section. Both the transmitter and receiver sections comprise conventional telescope designs. Laser light emitted by the transmitter strikes a surface some distance away. A fraction of the light scattered from that surface is collected by the receiver's primary mirror and concentrated onto the photodetector. The optical components are configured in a package that is approximately 35 cm long, 20 cm high, and 20 cm wide.

The laser source and receiver electronics were assembled in a simple wavelength modulation spectroscopy (WMS) configuration. A fiber-coupled telecommunications-grade distributed feedback (DFB) diode laser was powered by an ILX Model LDX 3620 Current Source. Its temperature was regulated with an ILX Model 5910B Temperature Controller. The laser package contains a thermoelectric cooler for temperature regulation, and an optical isolator to prevent feedback of reflected light.

To perform WMS, the laser temperature and injection current were set initially to provide a laser wavelength corresponding to an absorption line of the target gas. As illustrated by Figure 2, the waveform synthesizer was then used to modulate the injection current, and thus the laser wavelength, to cause the wavelength to periodically sweep back and forth across the absorption line. When target gas was present in the optical path, the power received at the detector was reduced each time the wavelength swept across the absorption line. Thus, the detected power contained an amplitude modulation at a frequency that is twice the wavelength modulation frequency. The amplitude of this so-called 2f signal was measured precisely with an EG&G Model 5208 lock-in amplifier, using the waveform synthesizer's reference signal as the phase reference input to the lock-in.

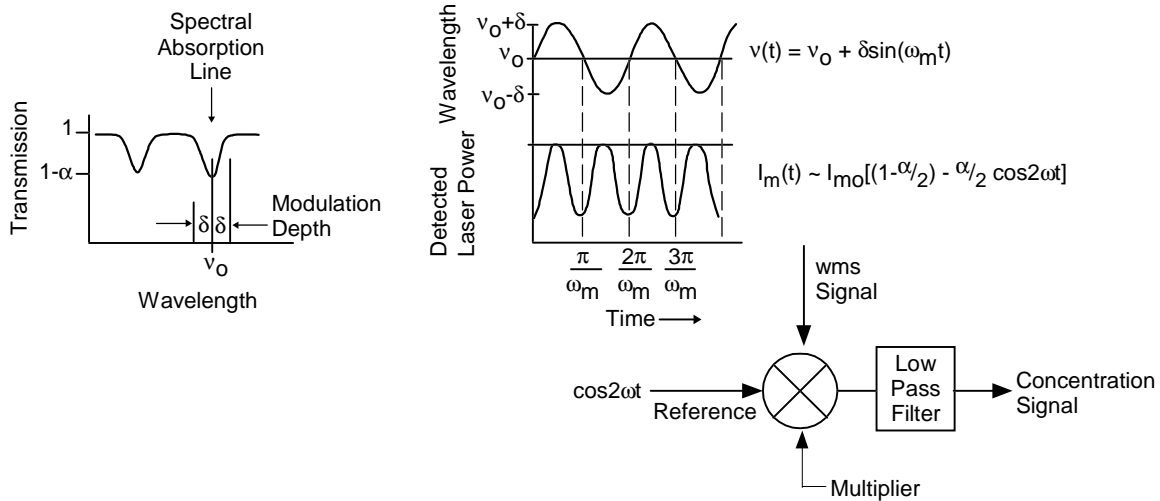


Figure 2. Basic wavelength modulation spectroscopy.

The output of the lock-in was a signal proportional to the path-integrated concentration of target gas between the laser transmitter and receiver. The signal is also proportional to the average power received by the detector, which as described above varies with the reflectance of the scattering surface as well as with the distance between the transmitter and the surface. Thus, a separate circuit was built to measure the average received laser power. This circuit was a simple low-pass filter followed by an amplifier. When measuring gas concentrations in the optical path, the ratio of $2f$ output signal from the lock-in amplifier divided by this average power signal can provide a value that is independent of received power. Little effort was made to optimize the selection of components comprising the low pass filter, and it became the limiting noise source in the sensor.

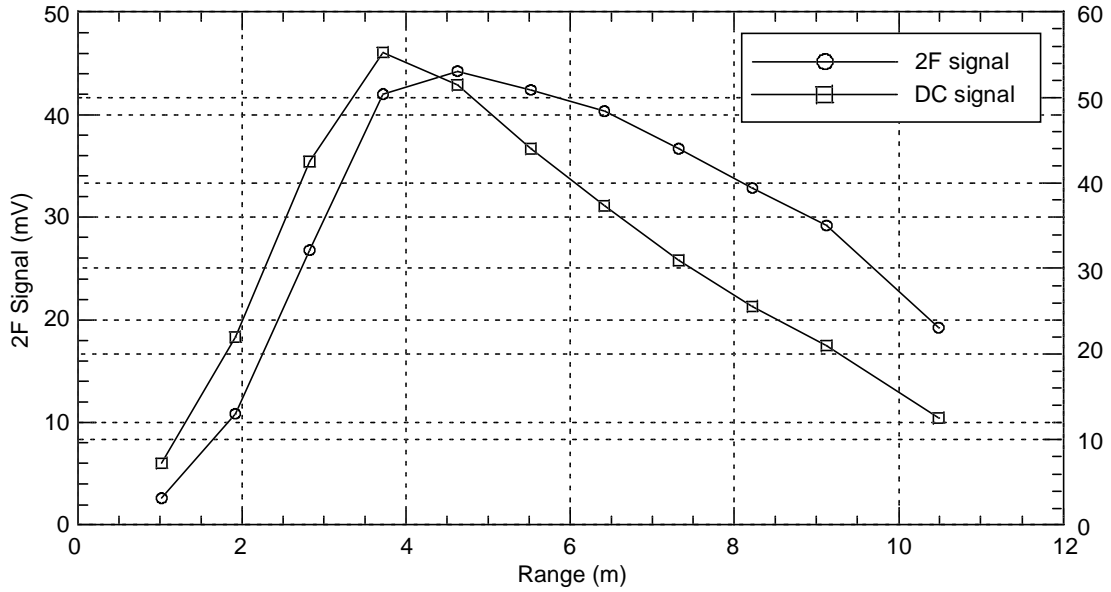
4. EXPERIMENTAL PROCEDURES AND DATA

The experimental work performed using the instrumentation described above confirmed the estimates of instrument performance. The experimental work comprised three steps: 1) measuring the detected optical power versus distance to the background; 2) demonstrating linear response to target gas concentration; and 3) determining the minimum measurable target gas concentration at a selected distance. The Noise Equivalent Spectral Absorbance (NESA) was determined from these three sets of measurements. The ability to detect a methane plume from a simulated pipe leak against a variety of backgrounds was also demonstrated.

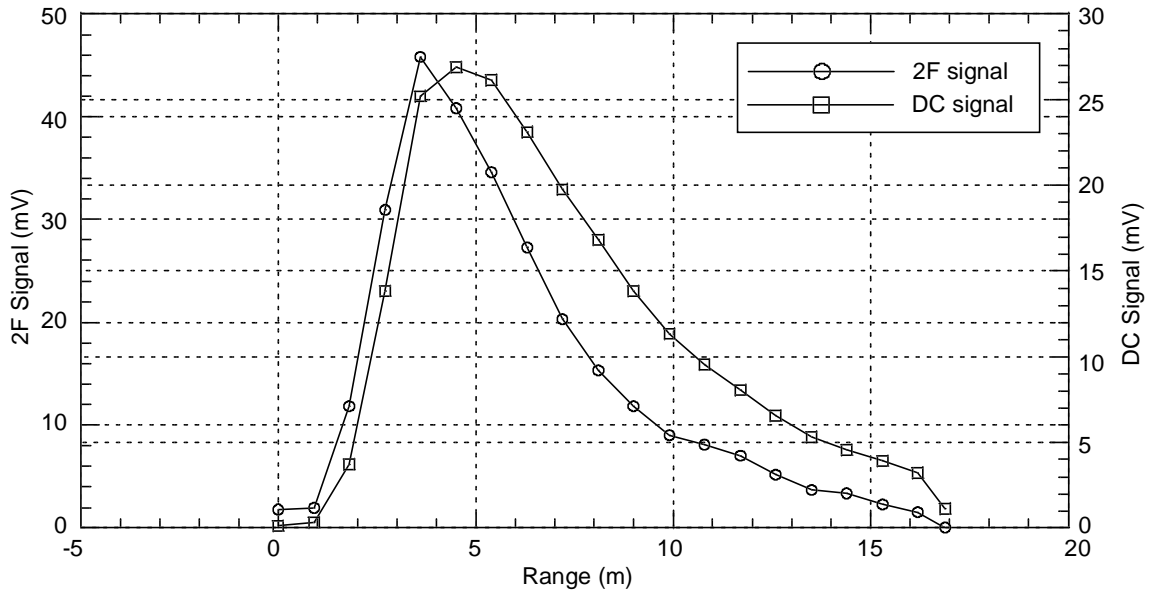
4.1. Optical power and linearity characterization

The performance of the optical system was initially evaluated using a fiber-coupled tunable diode laser having a nominal wavelength near $1.4 \mu\text{m}$, suitable for sensing atmospheric water vapor at several spectral lines of varying strength. Data were acquired by illuminating with the laser a movable target made from a $25 \times 25 \text{ cm}$ flat section of brown corrugated cardboard on a tripod. The target position was varied at regularly spaced distances between about 1 and 12 m from the transceiver, the range being limited by available space in the laboratory. The average received power signal and the $2f$ signal was measured at each position.

Figures 3a and 3b show the measured signal amplitudes as a function of separation between the transceiver and the target for two separate trials. In both cases, the average power signal, denoted as “DC”, shows a peak at a range of about 4 m and, as range increases, follows a $1/L^2$ trend. This performance was anticipated from the optical design. As described previously, the $2f$ signals result from the product of the path-integrated water vapor concentration in the air and the received laser power. Since water vapor is uniformly distributed over the measurement path length, its path-integrated concentration is directly proportional to path length. Thus, the ratio of the $2f$ to the DC signals should increase linearly with path length (for small spectral absorbance where the exponential term in Eq. (1) can be approximated by a line) and pass through the origin (except for any electronic offset). Figures 4a and 4b plot this ratio as a function of path length. The linear relationship is clear,



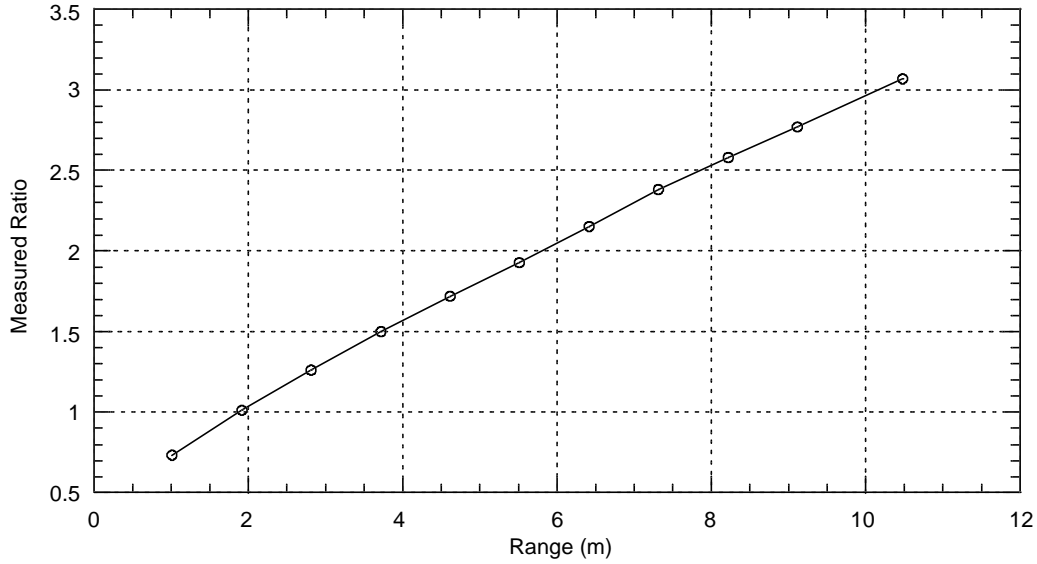
(a)



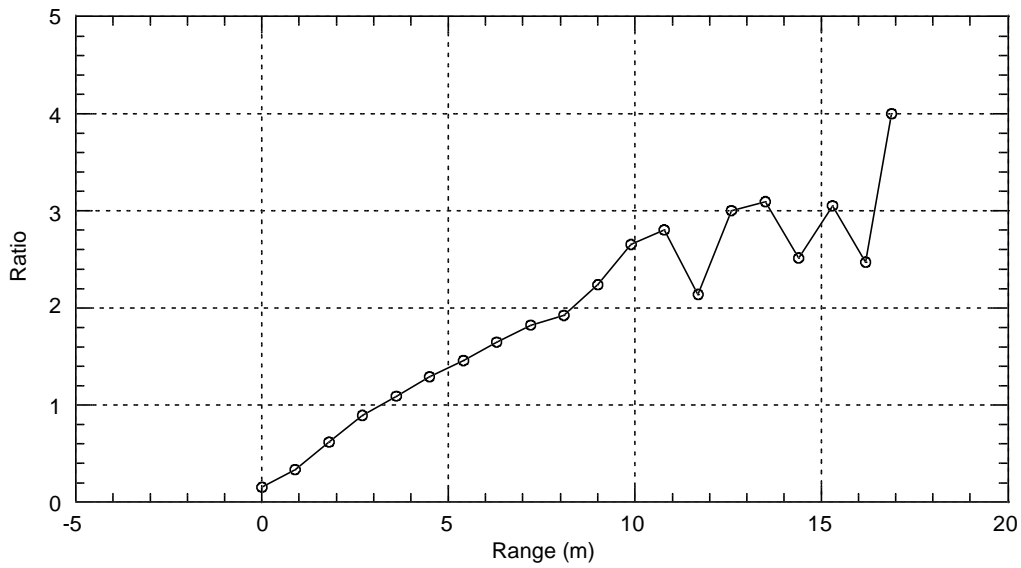
(b)

Figure 3. 2f and DC signals versus range with laser tuned to sense atmospheric water vapor.

especially in Figure 3a. (Slight deviation from linearity can be seen at the longer pathlengths due to high absorbance). Figure 4b shows the effect of noise in the DC signal at longer path lengths, resulting from sub-optimum design of the low pass filter.



(a)



(b)

Figure 4. Ratio of 2f to DC signals from data of Figure 3.

4.2. Minimum measurable concentration

A precision system calibration, and evaluation of the minimum detectable concentration, was performed using a cylindrical, calibrated flow cell with transparent windows on each end. The cell was placed within the optical path between the transceiver and laboratory wall. The distance between the transceiver and the wall was 4 m. A pair of mass flow controllers precisely regulated a mixture of methane and nitrogen that flowed through this cell. The maximum flow rates were 100 and 5,000 ml/min for the two gases respectively. Data were acquired by setting the flow ratio initially to 0.04, decreasing the ratio to zero, and then increasing it back to 0.18. Some hysteresis was observed due to insufficient mass flow controller warm-up. The optical path length through the cell was 12 cm, so the ratio (in ppm) of methane flow rate to nitrogen flow rate multiplied by 0.12 m yields the path-integrated concentration of methane in ppm-m.

Figure 5 plots ppm-m versus the 2f signal. The slope of 22 mV/ppm-m is the so-called “calibration constant” that relates the path-integrated concentration to the measured 2f signal. The intercept of -106 ppm-m is the electronic offset.

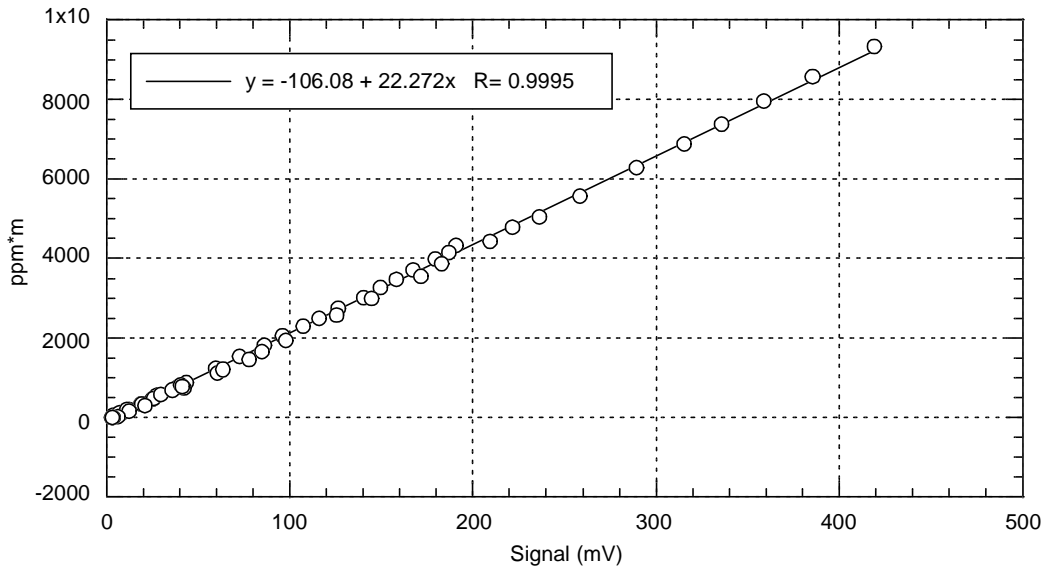


Figure 5. Calibration.

The minimum detectable concentration was determined from the statistical fluctuations over time of the measured 2f signal with fixed methane concentration. Three values of equivalent methane concentration were chosen for this measurement: 0 ppm-m (representing the natural background of methane in the laboratory air), 211 ppm-m, and 1152 ppm-m. For each concentration, 600 data samples were acquired at a rate of one per second. The data are plotted in Figure 6. The rms noise is seen to be essentially independent of concentration, with an average value of about 0.39 mV, corresponding to about 8.5 ppm-m, or a NESAs of 8.5×10^{-5} . Thus, the detection limits using this breadboard system are about 8.5x the values listed in Table 1.

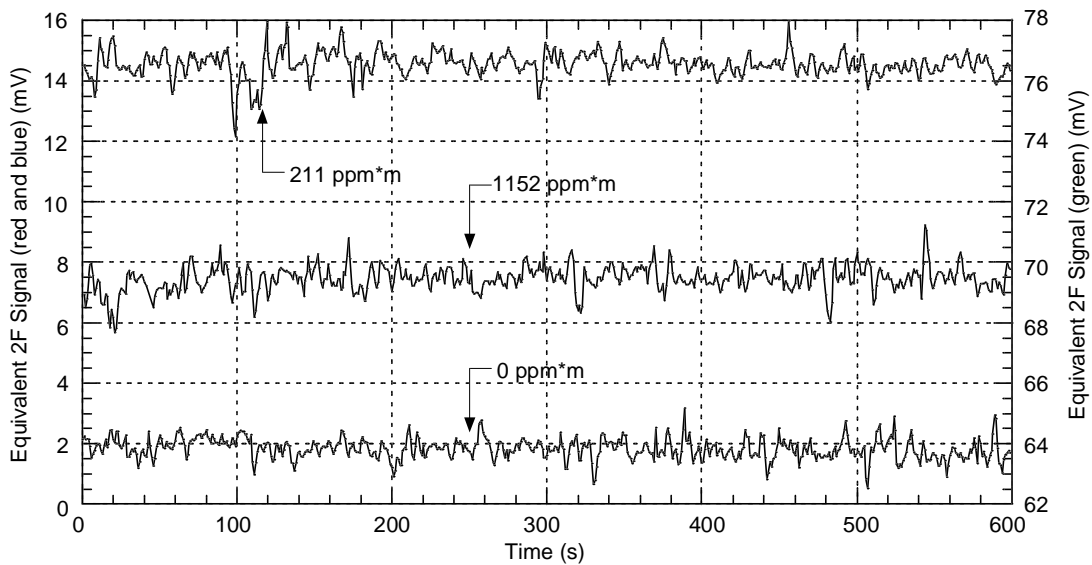


Figure 6. Temporal history of 2f signals observing fixed methane concentrations.

Figure 7 shows that when the range to the scattering target is changed, but methane concentration is held constant, the magnitude of the noise changes roughly in proportion to the signal. These data indicate that the primary noise source is not in the receiver electronics, as in that case the noise would be independent of signal. Instead, the data suggest that the dominant noise source is in the transmitted laser beam. Experience shows that this noise can be reduced by nearly an order of magnitude with improved engineering.

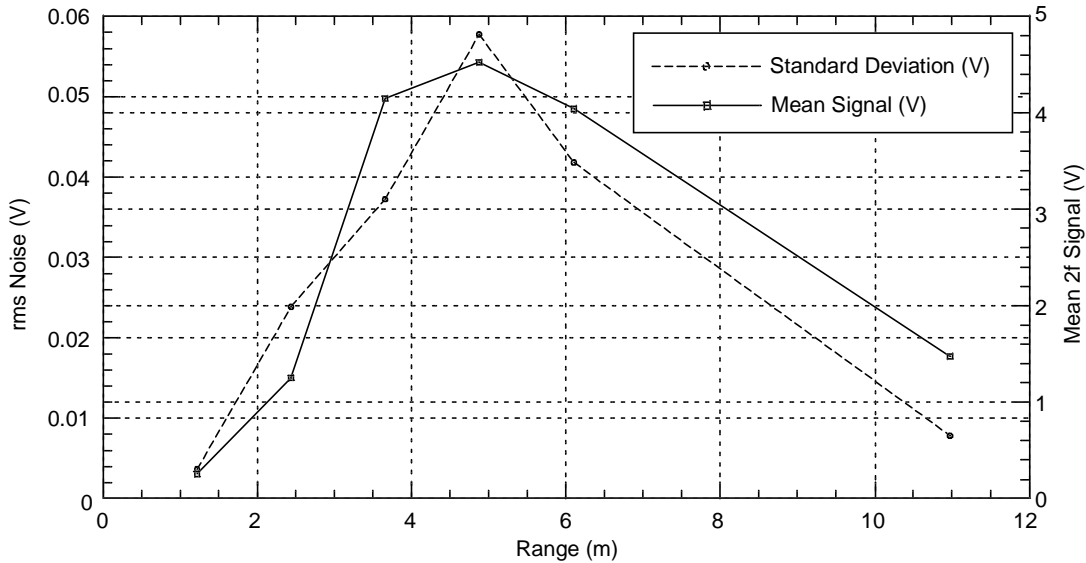


Figure 7. Noise versus range for fixed methane concentrations.

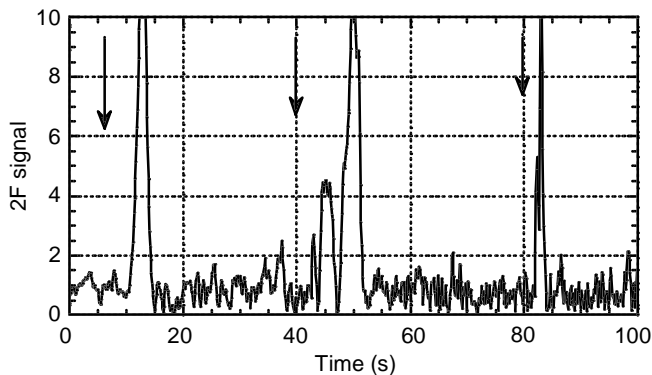
4.3. Outdoor field testing

To demonstrate an ability to detect simulated methane leaks from pipes, the TDLAS sensor, the data acquisition system, and methane gas cylinder were placed mobile carts and rolled outside. The methane cylinder was attached to a hand-held welder's nozzle which could be opened and closed quickly. Connection of the nozzle directly to a mass flow meter showed the typical flow rate to be between 5 and 7 lpm.

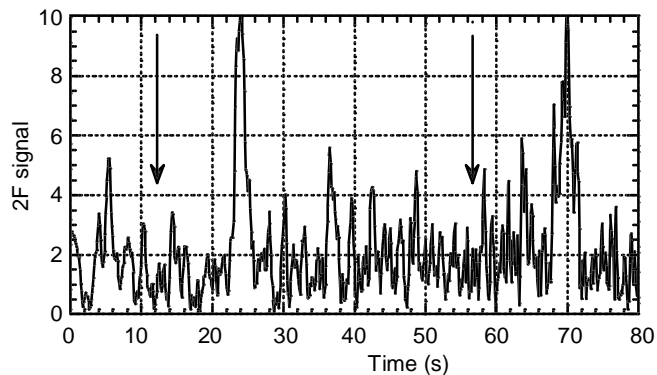
Five methane leak measurements were conducted on a windy day, using various available topographic targets as the scattering surfaces. The procedure for the measurement was essentially the same for each of the first four trials: 1) a topographic target was selected and the leak sensor's laser beam was directed towards that target; 2) the methane leak source nozzle was placed in the vicinity of the inspection site, the acquisition program was initiated and computer recording of the received 2f signal began; 3) the methane leak was initiated by opening the nozzle; 4) when the leak was detected in the 2f signal, the nozzle was closed; 5) when the 2f signal subsided, the nozzle was opened again until several leaks had been detected. Figures 8a-d show the results, indicating the ease of detecting gas against a variety of backgrounds.

5. CONCLUSION

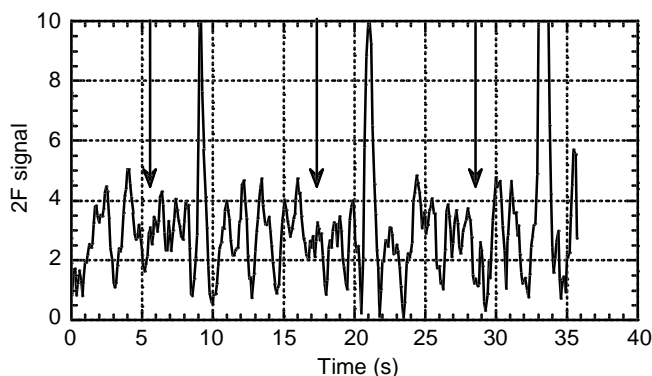
A hand-held TDLAS system that projects a laser beam onto an unremarkable topographic target up to 20 m away, receives laser light backscattered from that target, and deduces the concentration of a specific gas within the optical path has been demonstrated. It provided a noise equivalent spectral absorbance of about 8×10^{-5} , with the dominant noise source being optical noise in the transmitted laser beam. This provided a capability to detect a path-integrated concentration of methane less than 10 ppm-m in 1 second with unity signal-to-noise ratio. The ability to use the device to unambiguously detect gas leaks from point outdoor sources was illustrated.



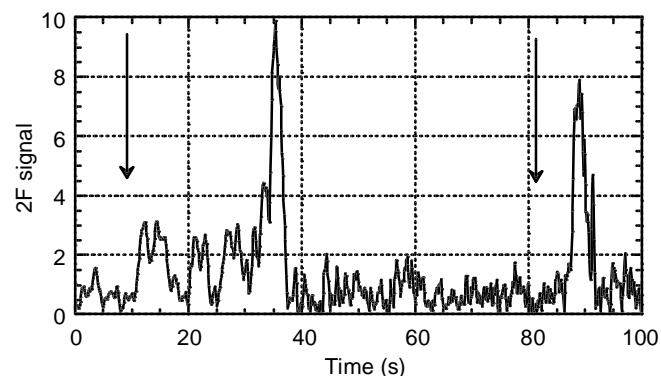
(a) 12 ft from a brick wall



(b) 24 ft from a brick wall



(c) 22 ft from a grassy embankment



(d) 17 ft from a hedge

Figure 8. Simulated gas leak detection. Arrows indicate nozzle opening.

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