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DEVELOPMENT OF FREQUENCY-AGILE  
HIGH-REPETITION-RATE CO2 DIAL SYSTEMS  
FOR LONG RANGE CHEMICAL REMOTE SENSING

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# Development of Frequency-Agile High-Repetition-Rate CO<sub>2</sub> DIAL Systems for Long Range Chemical Remote Sensing

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## Abstract

Issues related to the development of direct detection, long-range CO<sub>2</sub> DIAL systems for chemical detection and identification are presented and discussed including : data handling and display techniques for large, multi- $\lambda$  data sets, turbulence effects, slant path propagation, and speckle averaging. Data examples from various field campaigns and CO<sub>2</sub> lidar platforms are used to illustrate the issues.

**Keywords:** CO<sub>2</sub> DIAL, laser remote sensing, chemical detection, lidar, airborne

## Introduction

There is a vast literature describing the advances in DIAL lidar technology that have occurred over the last twenty years. Much of the early work is discussed and referenced in the books by R. Measures<sup>1</sup>, and E. Hinkley.<sup>2</sup> The DIAL technique itself has been critically reviewed by Zanzoterra.<sup>3</sup> An extensive collection of LIDAR references in general can be found at a web site<sup>4</sup> organized by W. Grant. Much useful information pertinent to CO<sub>2</sub> DIAL applications can be found in the book by A. Jelalian<sup>5</sup>

A variety of chemicals have been reported as detected in the open atmosphere using CO<sub>2</sub> DIAL. Some examples of particular chemicals include : H<sub>2</sub>O<sup>6,7</sup>, SF<sub>6</sub><sup>8</sup>, C<sub>2</sub>H<sub>4</sub><sup>9</sup>, O<sub>3</sub><sup>10</sup>, NH<sub>3</sub><sup>11</sup>, and various hydrazine<sup>12</sup> and organophosphate<sup>8</sup> compounds to name a few. Many other chemicals have appreciable absorption in the 9-11  $\mu$ m region. In a recent example, SF<sub>6</sub> was detected at ~16 km range with a ground based system using mountains as the backscatter target.<sup>8</sup> Much larger detection ranges can be accomplished through the use of retro-reflector technology, which can yield a return signal many orders of magnitude larger than topographic targets. Several groups have previously demonstrated airborne operation of CO<sub>2</sub> DIAL lidar systems.<sup>10,13-16</sup>

There has been much discussion in the literature describing the various sources of error in CO<sub>2</sub> DIAL measurements and the resulting pulse averaging statistics.<sup>17-28</sup> External factors to consider include the effects of differential surface albedo (reflectance), speckle, black-body radiation, atmospheric extinction, and atmospheric turbulence effects on the outgoing and return laser light such as beam spread, wander, and scintillation. Additional system factors to consider include detector noise, linearity and surface uniformity, laser beam mode, frequency, amplitude and pointing stability, amplifier noise, platform vibration and thermal stability, and overall data acquisition robustness.

A key factor in determining overall system chemical concentration detection limit (the differential absorption noise floor) is the signal averaging behavior of the return signal at each wavelength. Ideally, one would like the unavoidable return signal shot-to-shot fluctuations to exhibit a completely random behavior (i.e. no uncorrectable drifts in the mean) during the measurement time so that S/N will scale as the square root of the number of shots averaged together. In practice, there is

always a limit as to how random a data set is. Signal fluctuations that affect all  $\lambda$ 's the same way can be removed by normalizing the signal from one  $\lambda$  to another. Invariably, variations appear in the data set for each wavelength that cannot be removed by averaging or by taking the ratio. Differential absorption noise floors slightly less than 1% have been reported in the literature for a 25 shot average of pulse pair ratio data.<sup>27</sup> High repetition rate CO<sub>2</sub> lasers offer the possibility of quickly obtaining large data sets which will enable a more detailed examination of the limitations in pulse averaging statistics and the corresponding chemical concentration detection limits.

The design of an optimum CO<sub>2</sub> DIAL lidar system is influenced by such factors as desired range and sensitivity, limitations of the platform, available measurement time, etc. Technical issues related to the development of frequency-agile, high-repetition rate systems are discussed in the material that follows. The use of a moving airborne platform introduces some significant differences into the DIAL data sets as compared to ground-based systems which use a stationary backscatter target.

#### High-repetition, multi- $\lambda$ operation data sets

Most of the CO<sub>2</sub> dial reported in the literature has utilized the traditional two line method of determining the concentration of particular gas.  $\lambda_1$  is selected to be "on-resonance" with a spectral absorption feature for the species of interest, and  $\lambda_2$  is selected to be "off-resonance." This arrangement is adequate when one knows a priori that the difference in absorption between the two  $\lambda$ 's is due entirely to the species in question. However, the possibility of numerous unknown chemicals being present, with potentially overlapping absorption spectra, requires that the lidar system be capable of producing many more wavelengths. Thus, the available measurement time is likely to be split among all the different wavelengths emitted by the laser. So the challenge to the laser developer is to provide frequency-agile and high-repetition rate systems. In the case of CO<sub>2</sub> lasers, numerous schemes have been developed to increase the wavelength selection rate. Rotating mechanical methods have been demonstrated at repetition/selection rates >100 lines in 3 ms (>33 kHz) within a single burst, with burst rates ~ 360 Hz.<sup>29</sup> More recently, acousto-optic technology is being used to attempt tuning rates in excess of 100 kHz.<sup>30</sup> CO<sub>2</sub> lidar systems producing multiple wavelengths at 200 Hz repetition rate have been demonstrated.<sup>15,31</sup>

The flood of data that can result from such multi- $\lambda$  high repetition rate systems raises several data acquisition, display, storage, and analysis issues. The data sets will be essentially 3-dimensional arrays, where one axis indicates laser wavelength, a second axis indicates time or laser shot number, and the third axis would indicate the return signal amplitude. By normalizing the array to one particular  $\lambda$  (or the average of a group of  $\lambda$ s), a differential absorption surface can be generated (relative to the normalizing  $\lambda$ ). In the absence of knowledge that a given  $\lambda$  can be said to be "off-resonance" for all possible chemicals, such a surface cannot be said to be unique. The "noise" on the surface defined by these variables can be an important factor in establishing the minimum detectable absorption of the system. The minimum detectable gas concentration for any given species will depend not only on the noise on this surface but also on the presence of other chemicals that may cause interference. The techniques for analysis of infrared absorption spectra of chemical mixtures (sometimes referred to as chemometric analysis) to determine identity and concentration is an active area of investigation, particularly by those investigators developing Fourier Transform Infrared Spectroscopy (FTIR) based applications. Analysis of CO<sub>2</sub> lidar data with these methods has been described elsewhere.<sup>32</sup> In general, it is not only the noise in a particular  $\lambda$  that is important, but the overall noise in each multi- $\lambda$  absorption spectrum as well.

#### Turbulence Effects

Atmospheric turbulence produces a variety of effects of interest to lidar system development. Laser beam propagation in the atmosphere has been treated in many books<sup>33</sup> and is the subject of continuing investigations. Basically, the refractive index structure of the atmosphere modifies the laser light passing through it in various ways. The laser beam spot at the target can wander, increase in diameter, and acquire substantial amplitude and phase modulation (scintillation). Shot-to-shot

fluctuations will result if the target albedo (reflectance) varies on the scale of the laser spot structure (i.e. turbulence induced albedo fluctuations). One can also anticipate that the backscattered speckle field will be altered. First, by the fact that the spot pattern on target itself is different, and, second, by propagation back through the turbulence on the way to the detector.

### Slant path beam propagation through the atmosphere

An airborne lidar system looking and lingering on a particular point on the ground will have to propagate a laser beam at slant angles through the atmosphere. The range to the target during data collection will vary. Range variations can be large or small depending on the aircraft flight pattern. Large range variations will result if a single straight-line path past a site is used. Smaller range variations will result when the aircraft executes an orbit around the site while trying to maintain constant range. The  $\lambda$ -dependent extinction properties of the atmosphere throughout the data set will be a factor in determining how small a differential absorption can be observed. The laser energy on target and the backscattered return signal will be affected by temperature and concentration gradients and variations in the atmosphere throughout the data acquisition period. The most important molecular absorptions in the 9-11  $\mu\text{m}$  region are due to  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{O}_3$  and the continuum absorption. Aerosol extinction effects can also be important under heavy aerosol loading conditions or when clouds are present along the line-of-sight. Fig. 1 shows a calculation<sup>35</sup> of the slant path atmospheric transmission (U.S. Standard Atmosphere) at an altitude of 3.42 km in the 9 to 11  $\mu\text{m}$  region, with a round trip distance of 20 km ( $20^\circ$  down look angle). Some of the signal loss due to  $\text{CO}_2$  absorption can be reduced by shifting the laser lines away from the atmospheric absorption by, for example, operating the  $\text{CO}_2$  laser with isotopic gas mixtures such as  $^{13}\text{C}^{16}\text{O}_2$ ,  $^{12}\text{C}^{18}\text{O}_2$ , and  $^{13}\text{C}^{18}\text{O}_2$ .  $\text{CO}_2$  lasers can also generate substantial power levels operating in "hot band" transitions<sup>34</sup> which produces substantial wavelength shifts. Isotopic laser operation also increases the number of chemicals amenable to detection.

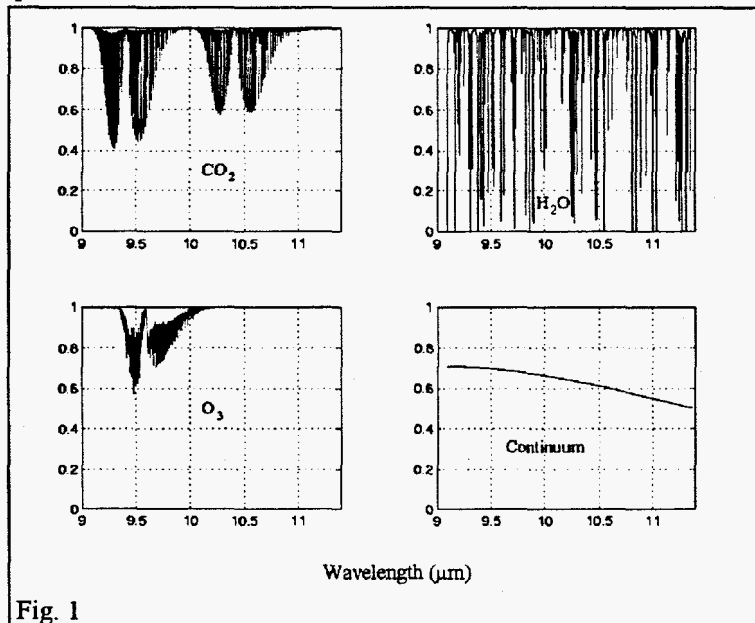


Fig. 1

To zeroth order, range variations during the data collection period affect the return signal in two ways. First, the detected signal falls off as  $1/R^2$  where  $R$  is the range to the target. Second, signal reduction due to atmospheric extinction is a path and wavelength dependent process. To illustrate the atmospheric extinction effect, Fig. 2 shows a calculation<sup>35</sup> of the difference in absorption that occurs when an airborne lidar at an altitude of 3.42 km changes its range-to-target from 10 km ( $20^\circ$  look angle) to 11 km ( $18.1^\circ$  look angle) while maintaining constant altitude. Attempting to use the data obtained at 10 km as

a "background" for data obtained at 11 km would produce the differential absorption results shown in Fig. 2. Continuum absorption causes changes on the order of 4-6%.  $\text{CO}_2$  absorption will cause a laser line dependent change as high as ~8%, and  $\text{O}_3$  as much as 4%. Laser lines affected by accidental coincidences with water vapor absorption lines can experience rather large variations with range. Thus, range variations will produce systematic line-to-line variations in the measured return signal which, if uncorrected, will adversely affect the lidar system's minimum observable differential absorption. Accurate removal of these effects will require knowledge of the  $\text{CO}_2$ ,  $\text{O}_3$ , and  $\text{H}_2\text{O}$  concentration profiles with altitude as well as temperature and pressure information. The line-to-line  $1/R^2$  variations with range

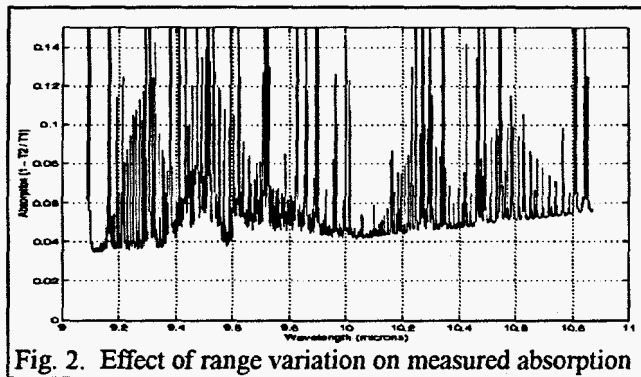


Fig. 2. Effect of range variation on measured absorption

can be mostly eliminated by measurement of the range for each shot, which can be obtained from the time-of arrival of the backscatter signal. Alternatively, one can simply allow the chemometric analysis of the absorption spectrum to include  $\text{CO}_2$ ,  $\text{O}_3$ , and  $\text{H}_2\text{O}$  as variables in the fit. However, the accuracy of a chemical concentration retrieved from chemometric analysis can depend on how many chemicals are being considered, so adding more can be detrimental.

### Speckle Averaging

Speckle noise refers to the spatial variations observed in the scattered field intensity when a highly monochromatic laser beam strikes a rough surface with features larger than the incident wavelength. The physics of speckle fields are well understood.<sup>36</sup> The shot-to-shot variability in the received signal by a small point sensor due to speckle can be as high as 100% (single speckle limit, standard deviation = mean), even if the return signal is very strong on average. The speckle size at the receiver is roughly  $\sim \lambda R/D$  where  $R$  is the range-to-target and  $D$  is a characteristic dimension of the laser spot on target. Shot-to-shot speckle noise in a lidar system is roughly inversely proportional to the square root of the number of speckles entering the receiver,<sup>36</sup> but only if each shot in the sequence produces a statistically independent speckle pattern. Once there is enough laser pulse energy so that the return signal fluctuations are dominated by speckle statistics, additional pulse energy does not improve the S/N any further. Under these conditions (speckle noise limit), one is better off using available laser power to generate more pulses and making additional, statistically independent measurements. For a stationary target, increasing the number of speckles can be done in many ways (although perhaps not very easily) including, for example, using a detection aperture that encompasses a great many speckles, reducing the size of the speckles at the receiver by increasing the laser spot on target, reducing the coherence length of the laser so that different parts of the target generate independent speckle fields during the pulse, or changing the amplitude and phase distribution of the laser beam on target between shots, or rotating the target. Atmospheric turbulence may play a part in varying the speckle field by modifying the amplitude and phase of the outgoing laser pulse as it travels towards the target. However, to obtain completely independent data from successive shots, one has to wait a sufficient amount of time for the atmosphere to change to the point where it does in fact generate a statistically independent laser field at the target. Laser pulses that are spaced too close to one another in time do not generate completely independent measurements.

For a moving platform with a sensor sampling many speckle in one shot, a new statistically independent measurement is obtained as soon as one moves  $\sim 1/2$  telescope diameter in distance perpendicular to the line-of-sight. Thus, an airborne system has the ability to sample a great many speckles as it flies through the speckle field. For example, for a 0.5 m diameter telescope traveling  $\sim 200$  m/s, it takes  $\sim 1.25$  ms. For example, a laser operating at  $10^4$  Hz scanning shot-to-shot over 50 different lines, the time between same  $\lambda$  pulses is  $\sim 5$  ms. For most target scenarios, the wavelength separation between laser lines is enough to assure independent speckle patterns when  $\lambda$  is changed. In this example, each individual wavelength will be averaged over independent speckle pattern realizations. Thus, an airborne platform has the ability to average over a great many independent target speckle fields.

### Experimental Results

One of the CO<sub>2</sub> DIAL platforms used is shown in Fig. 3. An optical layout diagram typical of this system is shown in Fig. 4. This layout applies to the data taken using lasers with a repetition rate limit of ~200 Hz. Typically the lasers were actually operated at only 10-20 Hz because they appeared to exhibit more stable, reliable operation which improved the quality of the data. Briefly, two separate CO<sub>2</sub> TEA lasers were made collinear with a polarization beam combiner. Laser beam divergence (>0.16 mrad) was adjusted with an off-axis beam expander. The expanded beam was made collinear with the optical axis of a 16" telescope and directed to selected targets with a large steering mirror. Initial measurements were taken with a 1 mm dia. HgCdTe detector cooled to 77° K with a cold bandpass filter. Typically, each laser could be instructed to emit as many as 44 different wavelengths in a user-selected pattern.

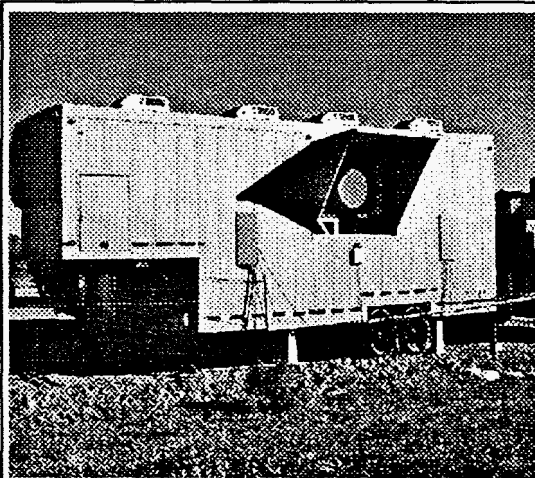


Fig. 3. Mobile CO<sub>2</sub> DIAL Platform

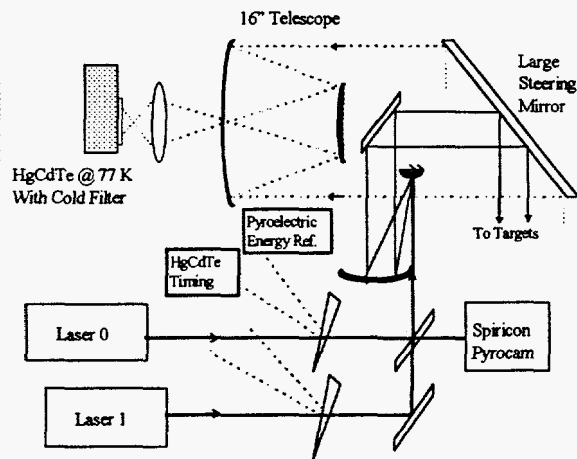


Fig. 4. Optical Layout

Various field campaigns have been conducted where chemical plumes, sometimes of known concentration, were interrogated at various ranges to examine system performance. Controlled chemical releases were produced with large wind tunnel where various chemicals, alone or in combination with others, were injected into the flow established by large fans. A variety of backscatter targets have been used in the measurements, including flame-sprayed aluminum, sand-blasted aluminum, plywood, concrete walls, tarp covered walls, and the mountain slopes in the background, as illustrated in Fig. 5.

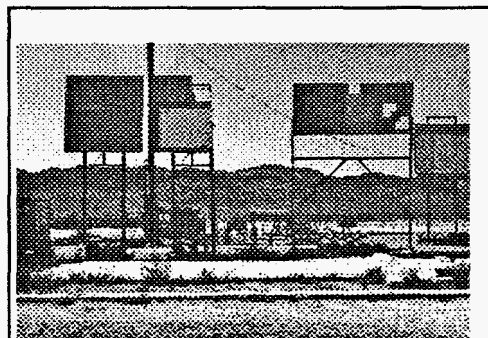


Fig. 5. Laser backscatter targets

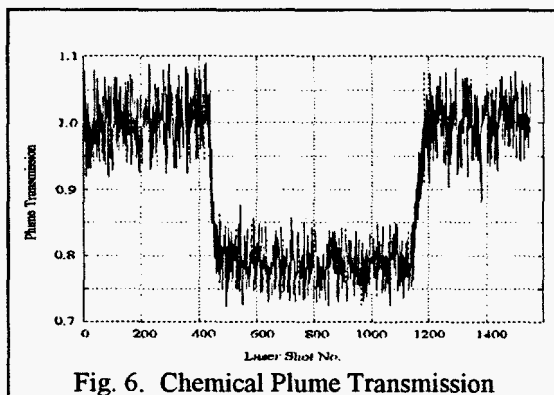


Fig. 6. Chemical Plume Transmission

One example of a controlled chemical release is shown in the transmission plot in Fig. 6. The sudden decrease in transmission observed at a single wavelength around laser shot 420 corresponds to when the chemical was released. The chemical was shut-off around laser shot 1150. This particular data set was obtained at a 500 m stand-off range using a FSA target, and exhibited shot-to-shot noise on the order of 3-4%. As discussed earlier, detecting and identifying particular chemicals with a high degree of confidence requires transmission (absorption) data sets containing many different



wavelengths, especially when the possibility exists that the plume under investigation contains many different chemicals with overlapping infrared absorption features. Thus, a DIAL laser system designer attempts to provide laser sources that can be rapidly tuned over a wide range of wavelengths, while maintaining good laser pulse energy, mode, and pointing stability. One example of a data set obtained using a 40- $\lambda$  scan pattern is shown in Fig. 7. The data is presented as a 3-dimensional surface where the three axis represent laser wavelength (or laser wavelength index number), time (or laser shot number), and absorption. In this particular data set, two different chemicals with non-overlapping absorption spectra were flowing at the start ( $t=0$ ) of the file. Approximately 300 s into the file, both chemicals are

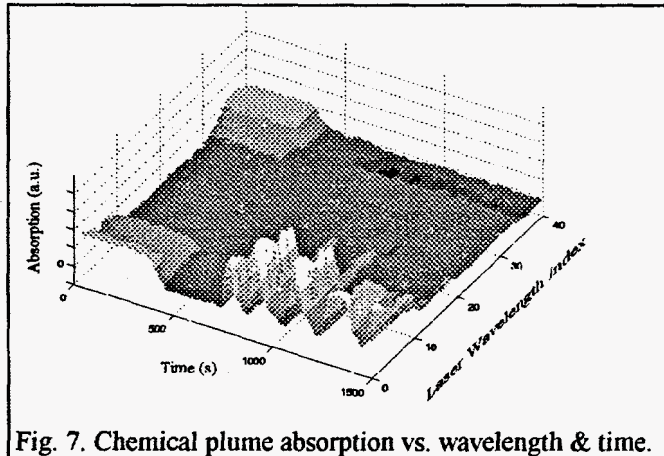


Fig. 7. Chemical plume absorption vs. wavelength & time.

shut off. Subsequently, a series of bursts of only one chemical took place. The "noise" on this 3-dimensional surface is an important system performance factor in determining how well a CO<sub>2</sub> DIAL platform can detect a particular chemical in the presence of others. This form of data presentation is also very useful to quickly determine systematic biases in the data acquisition. Results of chemometric analyses using least squares fitting of reference spectra to actual field data have been discussed elsewhere.<sup>32</sup>

Various measurements were made to attempt to quantify the effects of turbulence on the observed system performance. Measurements of the effective beam divergence (i.e. spot size at the target) were made by scanning the laser beam across a tall and slender object (like a telephone pole) as a function of turbulence level. A measure of turbulence level was obtained by making refractive index structure constant ( $C_n^2$ ) measurements throughout the day. The  $C_n^2$  variation throughout a 14 hour period spanned almost 3 orders of magnitude, as shown in Fig. 8. The lowest turbulence level occurred late in the early evening at  $\sim 7:30$  pm. The beam divergence data vs  $C_n^2$  are shown in Fig. 9. The lines in Fig. 9 show various laser beam propagation model predictions.

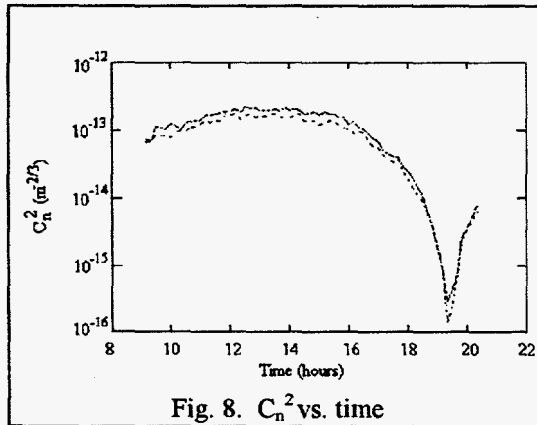


Fig. 8.  $C_n^2$  vs. time

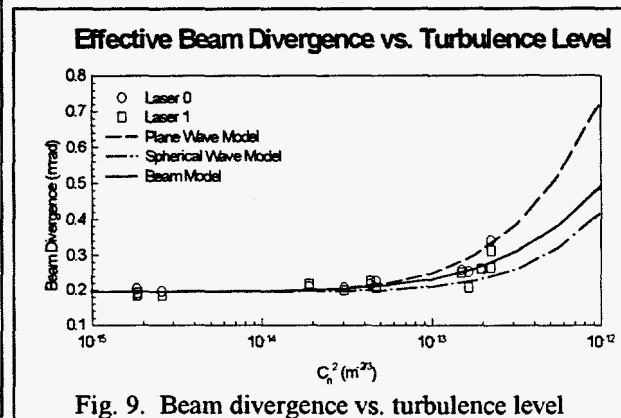


Fig. 9. Beam divergence vs. turbulence level

One example of the effects of turbulence on shot-to-shot noise levels is shown in Fig. 10 for the case of a 0.9 mrad beam striking a 8x12 foot FSA target at a range of 3.4 km. The RMS noise level for each of 20 different laser lines is shown for two different  $C_n^2$  turbulence conditions. The  $e^{-2}$  diameter of the beam is estimated to be on the order of  $\sim 3$  m, so that a significant fraction of the beam of the beam misses the high albedo FSA target. In this particular arrangement, it is likely that the lidar system is very sensitive to changes in laser spot patterns caused by turbulence.

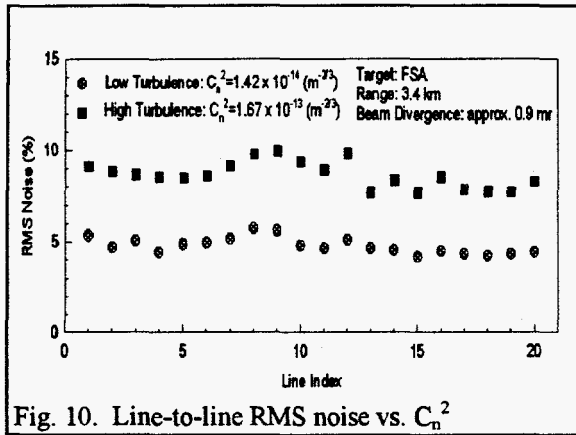


Fig. 10. Line-to-line RMS noise vs.  $C_n^2$

A preliminary examination of recent airborne CO<sub>2</sub> DIAL experiments provides a quick glance at some of the differences that can occur in the averaging statistics between a stationary platform and a moving one. Fig. 11 shows the pulse averaging statistics for two laser lines and their ratio as measured

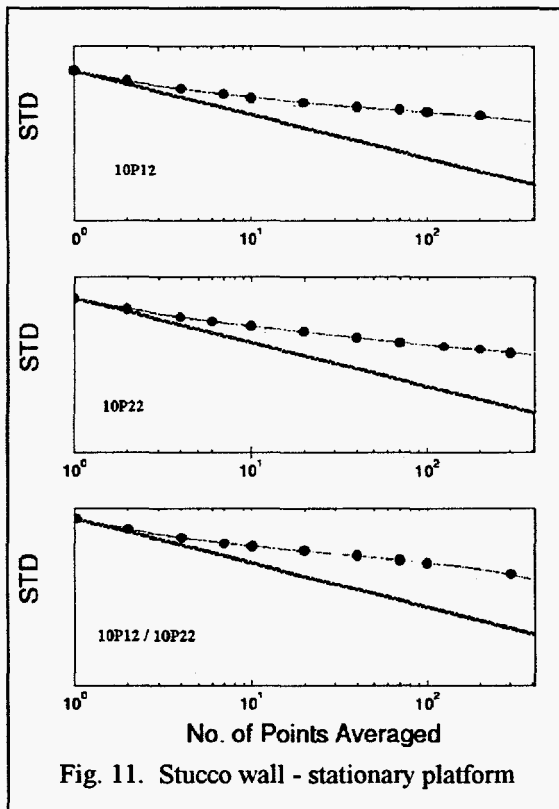


Fig. 11. Stucco wall - stationary platform

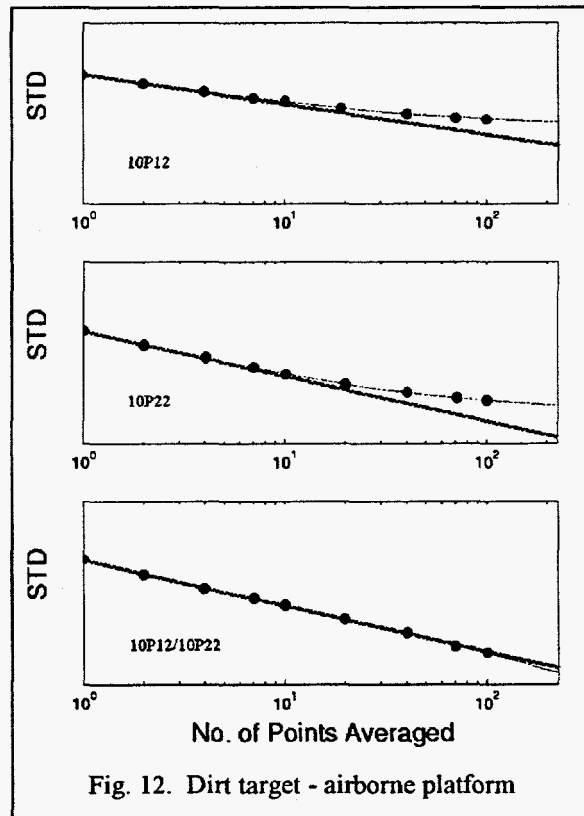


Fig. 12. Dirt target - airborne platform

from a stucco wall at close range while the platform was stationary. Fig. 12, shows the same two laser lines, but was obtained while the platform was airborne and aimed at a dirt target. The thick solid lines indicate the  $N^{1/2}$  behavior the data would follow if the data were completely random. Note that the airborne data for the pulse pair ratio follows a  $N^{1/2}$  dependence rather well, as compared to the equivalent data when the platform is stationary. This data suggests, but does not prove, that the airborne platform data may be averaging speckle field fluctuations more effectively than the stationary target.

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Metrolaser : K. Agar, et. al.

Additional LANL field team personnel participating in the early stages of the project include : G. Quigley, D. Greene

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