

# Balloon-borne tunable diode laser absorption spectrometer for multispecies trace gas measurements in the stratosphere

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A balloon-borne tunable diode laser absorption spectrometer capable of *in situ* measurement of the concentrations of several stratospheric trace gases simultaneously in the same parcel of air is described. Laboratory tests show present detection limits of 1.5 ppbv for NO and 0.5 ppbv for NO<sub>2</sub> under stratospheric conditions. This instrument was flown in July 1983 to make the first tunable diode laser measurement in the stratosphere. The concentration of NO<sub>2</sub> at 31.4 km and a solar zenith angle of 68° were measured to be  $13.4 \pm 5.2$  ppbv. The instrument was recovered intact. This success has led to plans to conduct additional NO/NO<sub>2</sub> flights and to upgrade the spectrometer to measure other gas species.

## 1. Introduction

Our understanding of stratospheric chemistry has increased markedly in the last decade. Much of this understanding has been gained from the interactive nature of reactive trace gas concentration measurements and predictions from photochemical models. The best test of these models is to compare predicted stratospheric concentrations to actual measurements. Because some measurements must be used as inputs to a model, a measurement of a single reactive species cannot be used as an independent test of that model. A set of concentration measurements of several reactive trace gases permits a much more rigorous test. Even though the literature describes a large number of reactive trace gas measurements, there are few simultaneous sets despite the fact that the concentrations are known to vary markedly with season, latitude, and time of day.<sup>1,2</sup> Thus existing measurements do not provide a satisfactory test of models. Even in cases where the data are obtained from a single platform, the nature of the instruments often means that the measurements are made on different air parcels or at different times. For example, on the Stratoprobe payload,<sup>3</sup> where measurement of NO, NO<sub>2</sub>, and HNO<sub>3</sub>

were made from a single platform, the NO altitude profile was measured *in situ* on ascent, the HNO<sub>3</sub> profile obtained was the average of a horizontal path of several kilometers, also measured on ascent, and the NO<sub>2</sub> profile was obtained by deconvolution of long path absorption measurements taken at sunrise or sunset. The preferred data for testing models are those which are gained by a set of instruments making multispecies concentration measurements on the same air parcel.

The use of high resolution IR spectroscopy for the measurement of stratospheric trace gas concentrations has a number of advantages over other methods. It is highly specific because at stratospheric pressures the linewidths of absorption lines approach the Doppler limit so that overlap of absorption lines from other species is unlikely. It is a universal technique that can be used for most small molecules. Because it can be used *in situ* and in real time without requiring the sample to be returned to the laboratory for analysis, it can be used to measure the concentrations of reactive species. The use of a tunable diode laser as the IR source adds high sensitivity to the advantages of IR spectroscopy. A tunable diode laser absorption spectrometer is thus a highly specific, highly sensitive, multispecies trace gas detector ideal for stratospheric applications.

This paper describes an *in situ* tunable diode laser absorption spectrometer (TDLAS) capable of measuring simultaneously the concentrations of several reactive species in the same air sample from a balloon platform. The TDLAS consists of a pair of lasers with cryogenic system, temperature control, and signal processing electronics. Also included are an onboard flow control and sampling system, a long path White cell, a

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concentration calibration system, and a command interface package. The TDLAS was built for the High Altitude Pollution Program (HAPP) as a joint project between Canada's York University and the United States' Perkin-Elmer Corp. Under a prior phase of the program, Perkin-Elmer had considered all possible measurement techniques and found that theoretically a TDLAS promised to best meet the requirements of a stratosphere measurement system.<sup>4</sup> The theoretical study was followed with a breadboarding stage<sup>5,6</sup> which confirmed the earlier recommendation by demonstrating in the laboratory sufficient sensitivity and specificity for studying the stratosphere. Finally, the breadboard was developed into the complete, ultrasensitive, and flightworthy spectrometer described in this paper.<sup>7</sup> Perkin-Elmer had the responsibility to design and build the entire optical system, the cryogenic system to house the lasers and the laser, temperature control, and signal processing electronics. York University was responsible for the flow control and sampling system, the onboard concentration calibration system, and the electronics to control these systems. The principal investigators of the flight phase are York University scientists. Finally, the University accomplished the integration of the subsystems into a balloon instrument and the interfacing of the mechanical and electronic packages to the balloon and to the balloon's command interface.

This instrument is presently configured to measure nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>) although it could be modified to measure the concentrations of other trace gases such as CO, HCl, N<sub>2</sub>O, CH<sub>4</sub>, or HNO<sub>3</sub> if satisfactory sampling and calibration procedures are established. It could also be upgraded to measure one or two of these species in addition to NO and NO<sub>2</sub>.

## II. Tunable Diode Laser Absorption Spectrometer

The operating principles of the TDLAS have been detailed elsewhere<sup>8-13</sup> so this discussion will be restricted to aspects related to this particular instrument and to stratospheric operation. The TDLAS technique is highly specific to the species under investigation, but to be useful in the stratosphere it must also be sensitive enough to make accurate concentration measurements. This requires the TDLAS to be able to measure absorptions of 10<sup>-7</sup>/m of path length. This sensitivity can be achieved either by using a path length of a kilometer or so with moderate absorption measurement capability or, as in this case, by using a multiple reflection White cell to give an optical path length of 48 m but requiring the ability to measure much smaller absorptions. This latter ability is accomplished by the use of frequency modulation techniques where the laser wavelength is modulated and the absorption is measured at twice the modulation frequency. This is the so-called 2*f* or second harmonic detection scheme originally used by Sachse *et al.*<sup>9</sup> and later independently by Reid *et al.*<sup>10</sup>

This particular instrument has been provided with a number of novel features not always present in other TDLASs to meet the constraints of balloon operation

and improve performance. The laser beams (up to a maximum of four) are combined optically to traverse the same air sample within a single White cell to ensure truly simultaneous measurements. The combined beam is split to give the effect of a dual-beam spectrometer where one beam passes through the sample and the other through cells containing high concentrations of the target gases. This latter permanent and unambiguous signal acts as a wavelength calibration and ensures detection specificity. Reflective rather than refractive optical elements have been used throughout because they allow the optical system to be aligned in the visible and are less likely to produce unwanted scattering. (The return of scattered radiation into the beam has been shown to be a dominant noise source in most TDLAS due to the interference fringes formed.<sup>10</sup>) Finally, the lasers are cooled with liquid cryogen because the normal closed-cycle coolers are too heavy and require too much power for a balloon instrument.

The instrument can be conveniently divided into four sections: (1) the optoelectronic and optomechanical section which includes the lasers, optics, and detectors; (2) the laser control and signal processing electronics; (3) the sampling, calibration, and flow control section which samples the ambient air through the White cell in a controlled manner and adds known amounts of the target gas for calibration; and (4) the master controller which includes all timing circuits and the command and data interface which connects the instrument to the balloon command interface pack. Each of these sections will now be discussed in detail.

### A. Optoelectronic and Optomechanical Section

The separation between absorption maxima for different molecules is usually greater than the tuning range of a tunable diode laser so it is generally not possible to use a single laser to measure the concentration of more than one trace gas. This fact and the requirement for simultaneous measurements of separate species require a separate laser for each molecule under study. The lasers are mounted in individual 1-liter liquid-helium-grade Dewars and cooled with liquid neon (boiling point 27 K). Liquid neon was chosen over liquid helium because of its much higher volume latent heat of vaporization (107 kJ/liter of liquid vs 2.5 kJ/liter for helium). Because 1 liter of liquid neon per laser lasts for some 15–18 h (which is longer than our proposed balloon flights), a source Dewar and cryogenic transfer system are not required, resulting in a substantial savings in system complexity and weight. Since the laser cannot then be run at temperatures below ~35 K, much of the tuning range of the laser is lost. This circumstance makes selection of the laser somewhat more difficult, but the convenience of the liquid neon more than outweighs this disadvantage.

The laser temperature is controlled by a servo-controlled heater referenced to a silicon temperature-sensing diode. The laser and its heater assembly are mechanically connected to the cold plate of the Dewar

through a thermal resistance selected to allow good temperature control ( $\pm 0.1$  mK) with a reasonable response time yet using low input power. We estimate that the heat transfer through this route is  $< 1$  W. Additional parasitic heat loads of slightly  $< 1$  W give a typical run time of 15–18 h. The pressure in the Dewar is held at 18 psia (125 kPa) by a pressure relief valve to prevent excessive boiling of the cryogen at the lower pressures encountered in the flight.

A schematic of the optical system is shown in Fig. 1. The radiation from each laser is collected by 2.54-cm

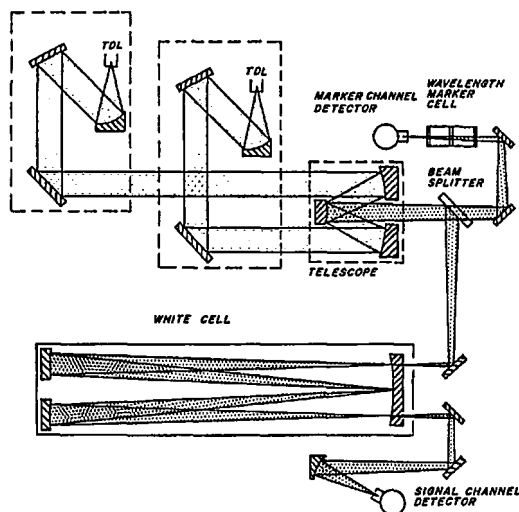


Fig. 1. Optical layout of the spectrometer.

(1-in.)  $f/2$  off-axis parabolic mirrors and collimated. The two 2.54-cm collimated beams then strike the primary of a telescope. The telescope focuses the parallel beams to a cone acceptable to the White cell and also combines the beams. Because the incoming beams are both parallel to the telescope axis, they are both focused at the telescope focus and then appear as off-axis cones within the  $f/30$  cone defined by the telescope. The foci for the two beams then remain coincident throughout the remainder of the optical system. Only two quadrants of the telescope primary are used, so the instrument can be upgraded to allow inclusion of two additional lasers. On exiting the telescope, the combined beam encounters a 90% reflecting beam splitter. The reflected beam is directed into a 1-m base path White cell so that the focus is at the field mirror. Inside the cell the beam traverses 48 m before exiting. It is then demagnified by an ellipsoidal mirror and focused onto a single signal detector. Because the foci of the two beams are coincident, a single  $1 \times 1$ -mm mercury cadmium telluride (HgCdTe) detector is used. The transmitted beam from the beam splitter is directed through cells containing high concentrations of the target gases to a single marker channel detector at the telescope focus.

To align the optical system, a Dewar is replaced by a mount containing a beam-expanded He-Ne laser focused to  $f/2$  to imitate the diode laser. Because the optical elements are reflective and the windows on the White cell and detector transmit in the visible, the optical train can be aligned using the He-Ne source. Once this alignment is accomplished, the diode laser Dewar assembly is returned, and the Dewar position is

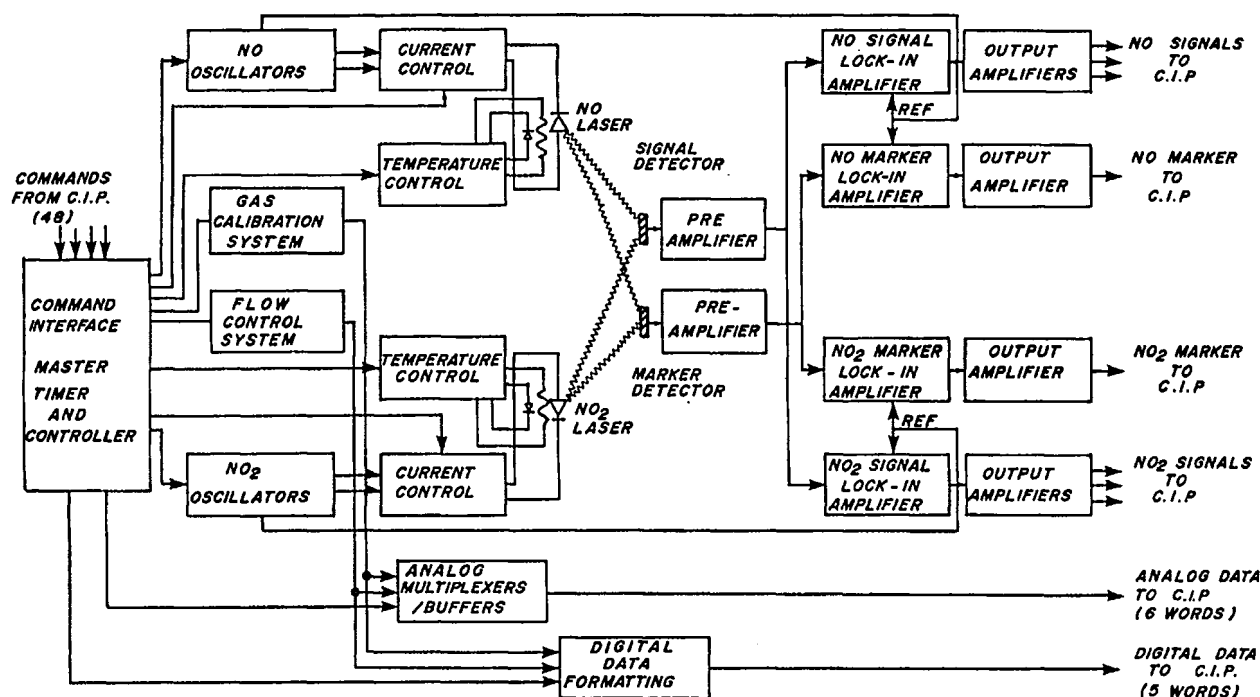


Fig. 2. Electronic block diagram.

adjusted to give the maximum power at the telescope focus.

## B. Laser Control and Signal Processing Electronics

An electronics block diagram including the master controller is shown in Fig. 2.

The current driving each laser has four components. The primary modulation component for the FM detection and an asynchronous modulation component to reduce the effect of interference fringes on the output are generated by oscillators and continuously fed to a modified Spectra-Physics Laser Control Module where it is added to the dc bias component which determines the base laser current and, therefore, the initial laser wavelength. Each laser has a different primary modulation frequency so that the contribution of each laser to the single detector output can be identified. Finally, a ramp waveform component (a low frequency, essentially dc component at  $3.33 \times 10^{-2}$  Hz) is used to scan the laser across the wavelength region of interest.

Only the ramp component is not permanently applied to the laser. On receiving a trigger pulse from the master timer, the current resets to the bias value and waits 1 sec to allow a start-of-scan pulse to be fed onto the signal and marker output lines, and then the ramp is applied. The ramp period is 30 sec, and then the ramp circuit resets and awaits another trigger pulse. When run off the internal timers, the trigger pulses are 32 sec apart. Thus there is  $\sim 1$  sec of dead time at the end of each scan.

The bias current can be adjusted up or down in flight to accommodate wavelength drifts in the laser output. The amplitudes of both modulations applied to a laser can be changed on the ground and in flight to continue to give an optimum signal as the pressure varies. A complicating factor is that the two modulations do not act on the laser completely independently, so a change in one necessitates a change in the other. The ramp circuit has three ranges selectable during flight so that the current can be scanned over a small range (about four linewidths) for the actual measurement or over

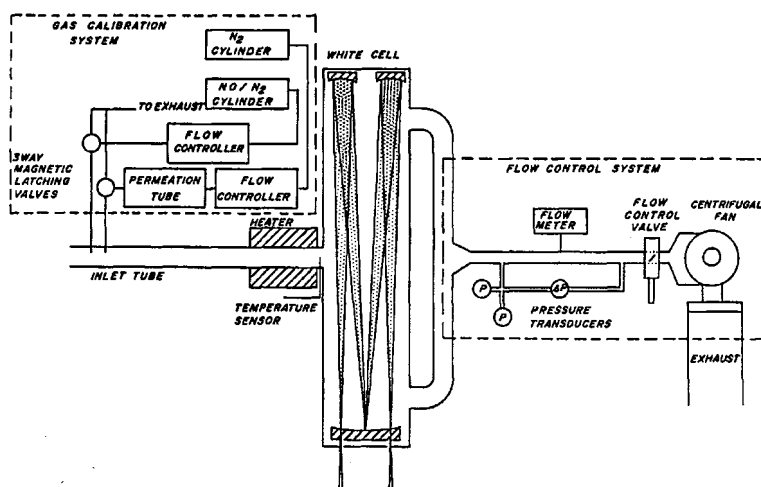
two wider ranges for setup and diagnostics. For example, if the laser wavelength drifts so that the small scan range no longer overlaps the absorption line, the line may be found using a wider ramp, and the bias current may be reset to locate the line within the small scan range. This feature can also assist in identifying any spectroscopic interferences from lines just outside the scan window. The ramp can also be directed to increase or decrease the current from the bias level.

The signals from the signal and marker detectors are preamplified by wideband amplifiers. Each preamplifier output is presented to two lock-in amplifiers that are referenced to twice the primary modulation frequency of the respective lasers. Because the lasers are modulated at different frequencies, the synchronous detection of each lock-in amplifier completely rejects the signal from the other laser so that the output signal is due to absorption of radiation from a single laser. The outputs of the marker lock-in amplifiers are scaled to make them compatible with the CIP digitizer, limited to prevent overload, and fed to the CIP. The outputs of the signal lock-in amplifiers are amplified by three separate amplifiers to give three output ranges whose relative gains can be set from the ground. These three outputs are scaled, limited, fed to the CIP, and sent to the ground station as separate words. There are eight output lines in all: two marker channels and six signal channels (three per gas).

## C. Sampling, Calibration, and Flow Control Systems

The sampling, calibration, and flow control systems are shown schematically in Fig. 3. For each trace gas measured, the calibration system will be different in terms of gas source and the materials that can be used. The present system which we describe here measures NO and NO<sub>2</sub>. The airflow is driven by a centrifugal fan which draws ambient air into the instrument via a heated glass inlet tube which is 122 cm (4 ft) long and 2.54 cm (1 in) in diameter. The air enters the glass-lined, stainless steel, White cell through a centrally located port and exits from two symmetrically located ports at the ends of the White cell diametrically oppo-

Fig. 3. Sampling, calibration, and flow control systems.



site to the inlet port. The two flows are recombined, and all the air passes two flowmeters, two pressure transducers (0–250 and 0–50 Torr), and a flow control valve before passing through the fan to be exhausted as far from the inlet as possible. The flow rate is such that the residence time in the inlet tube and White cell is <3 sec.

The calibration of signal against gas mixing ratio is accomplished by adding a known mixing ratio of each gas to the upstream end of the inlet tube. The total flow is monitored by two flowmeters. At high pressures (above 80 Torr), a mass flowmeter is used; but at lower pressures, the pressure drop down a length of straight tube is measured. Because both flowmeters are pressure dependent, they are calibrated in the laboratory over the pressure regime to be encountered during a balloon flight. A measured flow of between 10 and 50 standard  $\text{cm}^3/\text{min}$  (sccm) from a calibrated  $\text{NO}/\text{N}_2$  mixture cylinder of between 4 and 10 ppmv is continuously vented to the instrument exhaust via a three-way magnetic latching valve. For NO calibration this valve is opened to direct the  $\text{NO}/\text{N}_2$  flow into the inlet tube. A measured flow of 10 sccm of nitrogen is passed over a thermostated  $\text{NO}_2$  permeation tube to produce a  $\text{NO}_2/\text{N}_2$  mixture of 2–10 ppmv and is then handled identically to the  $\text{NO}/\text{N}_2$  mixture. The calibration system is made of stainless steel interconnected with stainless steel or Teflon tubing.

The mixing ratio of gas added to the White cell is calculated from the trace gas mixing ratio at its source, its flow, and the total airflow. The addition of approximately ambient mixing ratios of the trace gas to the air sample at the upstream end of the inlet tube ensures that any losses in the sampling system are the same for both the calibration gas and the sample gas. The measurements are thus compensated for any such losses. A problem that does arise is that there is a net gas phase production of  $\text{NO}_2$  from the reaction of NO with  $\text{O}_3$  once the air sample is removed from sunlight. The NO calibration compensates for the loss of NO, but the  $\text{NO}_2$  calibration cannot account for the production of  $\text{NO}_2$ . This information can be obtained, however, from the increase in the  $\text{NO}_2$  during the NO calibration. The NO and  $\text{NO}_2$  calibrations are performed sequentially rather than simultaneously for this reason.

The calibration and measurement cycles of the instrument are usually triggered by the pulse supplied to the ramp by the master timer. The calibration system logic counts the trigger pulses, and, on reaching a preset value, the valves are opened or closed within the 1-sec delay at the beginning of a scan. Since the residence time is of the order of a few seconds, the signal measurement on the next trace will be taken at nearly stable flow conditions. Laboratory tests have shown that the calibration signal stabilizes in, at most, one scan time (32–33 sec), although we calibrate for a minimum of four scan times in flight.

#### D. Master Controller

The instrument is designed to be compatible with the National Scientific Balloon Facility (NSBF) and National Research Council (NRC) of Canada Command Interface Packages (CIP). The two CIP packages have some differences in detail of operation but are conceptually the same. They can take analog or digital data from the instruments on the balloon, digitize it if necessary, format it, and telemeter it to the ground. The received data can be recovered in digital format or can be converted back to analog form to be compatible with recording requirements.

The signal channel outputs (three per trace gas) and the two marker channel outputs are fed in analog form to the CIP for digitization and transmission to the ground. The outputs from the pressure transducers, flowmeters, and temperature transducers along with analog monitor outputs and analog status information are multiplexed, either 8 or 16 outputs to a data line, and fed in analog form into the CIP. The status information that is inherently digital (counter values, valve open/closed status, and subsection on/off status) is formatted into 8-bit words and can be fed to the CIP either serially or in parallel as dictated by the CIP. At present, the original analog information is converted back from digital to analog form on the ground and recorded on a strip chart recorder. It is planned to have digital data recording in the future for easier data analysis. The digital words presently contain only status information for running the instrument in flight and are not recorded.

The instrument is designed to be run completely by its own timers. The master clock is either an onboard oscillator or the bit clock received from the CIP. This 32-kHz clock is divided down synchronously to provide all the required timing signals. The most important signal is the trigger pulse which is generated every 32–33 sec. This trigger pulse starts the ramp for each laser, increments the counters on the calibration system, activates the start of scan pulse on the signal and marker outputs, and synchronizes the analog data multiplexers.

Complete manual operation of the instrument in flight from the ground is possible using only the single-switch closure-type commands to override the onboard control. The commands are received from the CIP and (except for the power on/off commands) are optically isolated from the CIP and shaped before being fed to the instrument. The entire instrument can be powered up or down, and the lasers and heaters or the fan can be powered on or off independently. The trigger pulse to one laser can be disabled and the ramp activated independently of the master timer. The valve timer can also be overridden and the valves activated directly so that calibrations can be carried out at any time. The flow of the  $\text{NO}/\text{N}_2$  calibration gas can be varied to give three different NO concentrations in the White cell. The dc starting current to the laser can be incremented as can the direction and magnitude of the current ramp. The magnitudes of both modula-

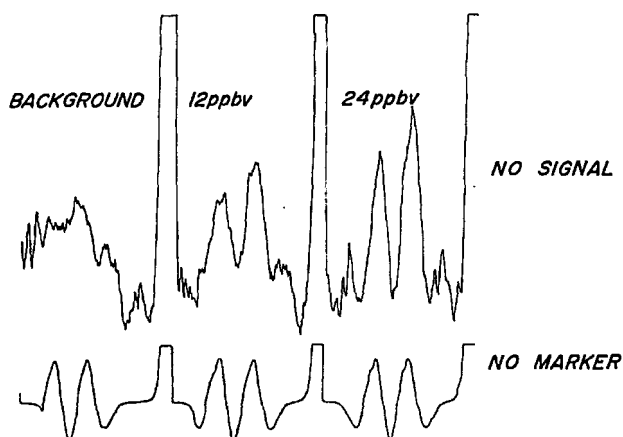


Fig. 4. Typical output traces of NO. The top trace is the NO signal channel, and the bottom is the associated marker channel.

tions can be varied up or down as can the laser temperature set point.

### III. Laboratory Performance of the TDLAS

A typical output of the instrument obtained in the laboratory is shown in Fig. 4. The upper trace is of one signal channel of NO, and the lower is its associated marker channel. The line under study is the  $R(6,5)$  resolved doublet at  $1896.984/1896.995 \text{ cm}^{-1}$ . These data are taken in synthetic air doped with varying amounts of NO. The signal trace also shows the background noise when there is no NO present.

We define the minimum detectable concentration (MDC) of the instrument as the concentration at which the ratio of peak-to-peak signal to peak-to-peak noise is unity. If we do not consider the background curvature as noise, these data show the instrument's MDC to be 1–2 ppbv for NO. Because the detection limit in terms of absorption is constant, we can estimate the detection limits for other species from their line strengths. Table I shows the detection limits of some stratospheric trace gases based on a limit of 1.5 ppbv for NO. It is evident that the instrument in its present form is capable of making stratospheric concentration measurements of CO, HNO<sub>3</sub>, N<sub>2</sub>O, CH<sub>4</sub>, and HCl in addition to NO and NO<sub>2</sub>.

The noise visible in Fig. 4 is frequency correlated rather than random and is a mixture of residual laser power fluctuations and fringe noise. The fringe noise results from scanning the laser wavelength across a resonant cavity inadvertently set up in the optical train. There appear to be a number of components to this noise. Some components can be eliminated by the asynchronous modulation. Residual noise from the incomplete removal of fringe noise from the White cell is visible but not limiting. The noise which presently limits performance originates in fringes which correspond to a path difference of a few tens of centimeters. These fringes are the most difficult to characterize because they fall within the signal frequencies, there appear to be several of them, and all are generated

Table I. Sensitivity of the TDLAS to Trace Stratospheric Species

Molecule	Approx. freq. (cm <sup>-1</sup> )	Line strength <sup>a</sup> (cm <sup>2</sup> -molecule-cm <sup>-1</sup> )	Estimated detection limit (ppbv)	Mixing ratio range (ppbv)
NO	1880	$6.0 \times 10^{-20}$	1.5	0.1–10
NO <sub>2</sub>	1600	$1.8 \times 10^{-19}$	0.5	0.1–10
HNO <sub>3</sub>	1720	$1.2 \times 10^{-19}$	0.8	0.1–10
N <sub>2</sub> O	1150	$6.5 \times 10^{-21}$	15.0	10–100
CO	2150	$4.4 \times 10^{-19}$	0.2	10–20
HCl	2900	$4.8 \times 10^{-19}$	0.2	0.2–2
CH <sub>4</sub>	1330	$5.6 \times 10^{-20}$	1.8	500–1000
<sup>b</sup> SO <sub>2</sub>	1370	$6.0 \times 10^{-20}$	1.5	0.05
<sup>b</sup> NH <sub>3</sub>	1065	$2.0 \times 10^{-19}$	0.5	0.2

<sup>a</sup> Ref. 14.

<sup>b</sup> Not potentially measurable.

before the beam splitter. We have had only limited success in removing these sources of noise.

Ultimately the detection limit of this instrument will be determined by the detector noise. The detectors have a noise equivalent power of  $3.3 \times 10^{-6} \mu\text{W}$  (3.3 pW). For the NO lines, the line strength  $S$  is  $6 \times 10^{-20} \text{ cm}^2\text{-molecule-cm}^{-1}$ . At 5 Torr and for a path length of 48 m, the noise equivalent concentration is  $0.3 \text{ ppbv}/\mu\text{W}$  of received power. The received power is  $\sim 10 \mu\text{W}$  so that the detector noise-limited MDC is 0.03 ppbv, nearly 2 orders of magnitude less than the observed MDC.

Although the present MDC for NO is only 1 or 2 ppbv compared with other reported values of 0.3 ppbv,<sup>10–13</sup> it is clear that the MDC can be substantially improved as the correlated noise is reduced before reaching the detector noise limit. We are taking two approaches to reducing this noise. First, we are identifying and eliminating the sources of scattered light. Second, we are planning to improve the data handling by incorporating digital data processing. This processing will allow postflight data manipulation including background noise subtraction and data averaging, both of which should improve the minimum detectable concentration.

The performance of the NO<sub>2</sub> side of the instrument does not yet match that of the NO side. The NO detection limit and the greater line strengths for NO<sub>2</sub> (see Table I) suggest that the detection limit for NO<sub>2</sub> should be 0.5 ppbv. Because the NO<sub>2</sub> laser does not operate in the regions where the NO<sub>2</sub> absorptions are at their maxima, we are using absorptions with line strengths about half of the maximum possible, and this doubles the expected detection limit to  $\sim 1 \text{ ppbv}$ . Experiments have shown the actual detection limit to be 1–2 ppbv, close to that for NO.

To check the performance of the instrument, a number of tests have been performed in the laboratory using synthetic air doped with known amounts of NO or NO<sub>2</sub>. The second harmonic signal is linear with concentration from the MDC up to hundreds of parts per billion as shown in Fig. 5. The response time of the second harmonic signal to changes in the concentration of the added gas is in the same order as the resi-

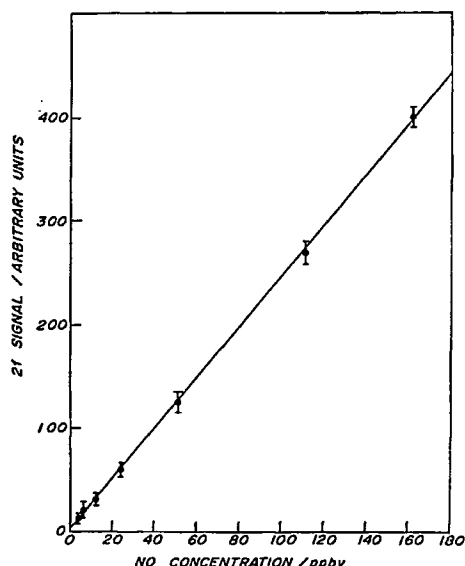


Fig. 5. Plot showing the linearity of the second-harmonic (2f) signal with mixing ratio. The error bars reflect only the precision in the measurement of the second-harmonic (2f) signal.

dence time, which shows that there is little or no retention of NO or NO<sub>2</sub> on the walls of the sampling system.

The instrument has been run for many hours in the laboratory, and, although there is a small laser wavelength drift (as would be expected from the large thermal resistance), the wavelength is easily tracked by the bias current increment commands. Otherwise, the circuits are stable over the pressure and temperature ranges expected in flight.

#### IV. Balloon Instrument: Configuration and Performance

For flight, the optical bench of the TDLAS is vibrationally isolated from the rest of the gondola. The bench as well as the sampling, calibration, and flow control systems and some electronics are enclosed in an insulated box thermostatically controlled to 25°C. The remaining electronics are uninsulated and mounted on the gondola to allow radiative cooling. The instrument is powered by lithium batteries, and the entire payload including the gondola, batteries, and a CIP pack weighs ~590 kg (1300 lb).

The instrument was flown from the National Research Council (Canada) balloon launch facility Gimli, Manitoba (50.62° N, 97.03° W) on 24 July 1983. It was lifted to 31.5 km by a  $1.06 \times 10^5$ -m<sup>3</sup> balloon for a total flight time of 8 h. Some noise problems were encountered in flight. On launch, a ground line was damaged, and it picked up a substantial amount of rf interference. This noise appeared on the analog signal outputs, and, because the CIP was being used to digitize the data, it also fed back into the instrument. The NO<sub>2</sub> laser appeared to be more susceptible to this noise than the NO laser, which is consistent with other observations of the relative stability of the two lasers. The engineering data, such as instrument tempera-

tures, pressures, flows, and the monitors on laser operation, battery power, and calibration mode, were extremely noisy but nevertheless sufficient for most analyses. The signal and marker channels were later filtered and some useful signals obtained. The NO marker signal was lost in the noise and never recovered. Absorption features were observed in the NO sample channel, and, although they could not be attributed to NO, they did show the wavelength stability of the instrument in flight to be better than specified. It was also determined that the optical system stayed in alignment throughout the pressure changes encountered. The introduced noise produced extreme wavelength drifts in the NO<sub>2</sub> laser, so it was not possible to observe a NO<sub>2</sub> absorption feature for more than a few scans. Using the signature in the NO<sub>2</sub> marker channel, NO<sub>2</sub> was identified in the White cell on two occasions. On the first, NO<sub>2</sub> was observed in eight successive laser scans. However, the instrument was in the NO<sub>2</sub> calibrate mode adding 10 ppbv of NO<sub>2</sub> to the ambient airstream. On the second occasion, one scan was taken during an NO<sub>2</sub> calibration and three scans during the measure mode. These scans allowed a calibrated measurement of NO<sub>2</sub>. The measured mixing ratio of NO<sub>2</sub>, at 31.4 km and a solar zenith angle of 68°, is  $13.4 \pm 5.2$  ppbv.

The instrument was recovered intact, checked, and made ready for a second flight, but a combination of support problems and bad weather resulted in the loss of the launch opportunity even though the instrument was operational.

#### V. Future Plans

The data obtained in flight have confirmed that the instrument is capable of making the measurements for which it was designed. The optics, the control electronics, and the sampling and calibration systems all functioned as expected through the flight. As far as can be ascertained in light of the pickup noise problem, there were no parts of the instrument which did not perform as expected.

The instrument will be upgraded slightly and flown again in the near future to make the simultaneous NO and NO<sub>2</sub> measurements. The upgrade will consist of digitizing the data before feeding it to the CIP and setting up a ground station to record the digital data so it can be readily analyzed by computer. Laboratory tests have shown that, in addition to making the analysis easier, computer averaging can substantially improve the detection limit of this type of instrument.

After it is used to measure NO and NO<sub>2</sub>, the instrument could be modified to measure other species such as CO, HCl, CH<sub>4</sub>, N<sub>2</sub>O, or HNO<sub>3</sub> or it could be further upgraded to measure three or four species simultaneously.

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