

# THE LASER ABSORPTION SPECTROMETER: A NEW REMOTE SENSING INSTRUMENT FOR ATMOSPHERIC POLLUTION MONITORING<sup>1</sup>

**M. S. SHUMATE**  
**Jet Propulsion Laboratory**  
**Pasadena, California 91103**

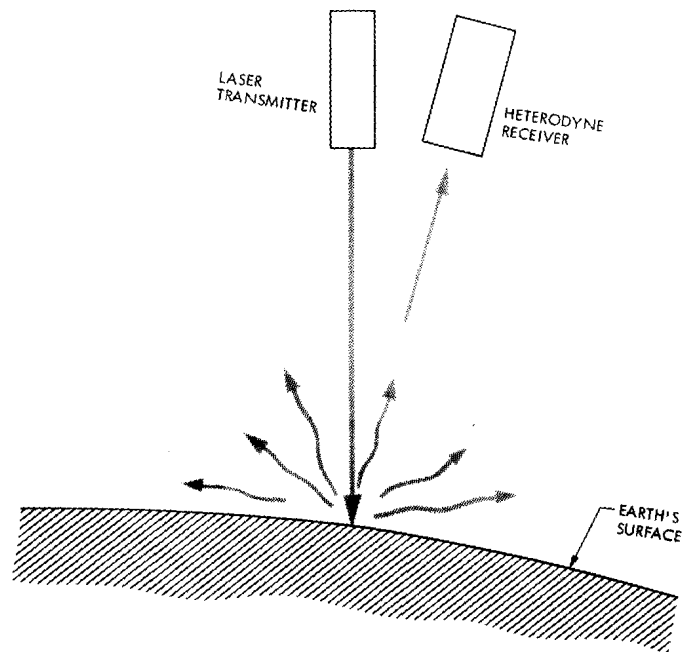
**Summary.** An instrument capable of remotely monitoring trace atmospheric constituents is described. The instrument, called a Laser Absorption Spectrometer, can be operated from an aircraft or spacecraft to measure the concentration of selected gases in three dimensions. This device will be particularly useful for rapid determination of pollutant levels in urban areas.

**Introduction.** This paper describes an instrument for monitoring the concentration of trace constituents in the atmosphere. The device utilizes infrared lasers, and is capable of measuring the amount of several important trace atmospheric constituents that constitute what is commonly called photochemical smog. The instrument is designed to operate from an aircraft, and will look downward toward the Earth, continuously measuring the concentrations of selected pollutants between the aircraft and the Earth below. An instrument using the same concept could also be designed to operate from satellite altitudes.

The principle of operation of this instrument, called the Laser Absorption Spectrometer (LAS), is depicted in Figure 1 (Ref. 1). The instrument contains an infrared laser transmitter and a heterodyne receiver which are capable of operation at several wavelengths. The laser signal is transmitted downward from the aircraft, and scatters off the ground. A small part of the reflected signal is detected by the heterodyne receiver. Operation at two different, closely spaced wavelengths will permit the determination of the amount of a specific pollutant in the optical path. One of the operating wavelengths is chosen so that it overlaps an absorption line of the pollutant molecule of interest. The second wavelength is chosen so that it does not overlap absorption lines of any variable atmospheric constituents. The latter wavelength serves to calibrate the losses on the path: scattering, surface absorption, and optical system. The former wavelength has an added

---

<sup>1</sup> This paper presents one phase of research carried out at the Jet Propulsion Laboratory, California Institute of Technology, under Contract NAS7-100, sponsored by the National Aeronautics and Space Administration.



**Figure 1. Laser Absorption Spectrometer Concept**

loss due to the absorption by the pollutant selected. The difference in the two signals provides a measure of the amount of the pollutant in the optical path.

An important feature of the LAS is that it is capable of measuring the concentrations of pollutants in the low-lying atmospheric layers. Since it is an active system, carrying its own controlled source, it does not depend upon scattered sunlight or thermal emissions for its operation. The system can be used any time of the day or night, and could produce a great deal of information about the spatial and temporal history of selected pollutants.

The successful operation of the LAS depends upon the coincidence between the emission line of an infrared laser and an absorption line of a specific pollutant. The pollutant line must be isolated from absorption lines of other atmospheric constituents, in order to provide an unambiguous measurement. Broadband sources and systems based on conventional spectrometers have limited usefulness due to the very narrow linewidths of most trace atmospheric gases in the middle infrared portion of the spectrum. Tunable lasers are available for use in the 2-10  $\mu\text{m}$  wavelength range (Refs. 2, 3) and have been used extensively to study the absorption properties of atmospheric gases (Ref. 4).

The tunable laser sources mentioned, infrared emitting diode type devices, have low output powers, and require cooling below 30 K for CW operation. Their tunability makes them ideally suited for laboratory use, and at limited fixed sites on the ground. For operation from an aircraft or spacecraft platform, higher laser powers are required, necessitating the consideration of other types of infrared lasers.

Several types of CW lasers that operate in the infrared are available. These lasers typically operate at vibration-rotation bands of polyatomic molecules, with an individual gas capable of producing laser oscillations at many different, closely spaced wavelengths. The most common infrared gas lasers use carbon monoxide and carbon dioxide, and operate in the 5-7  $\mu\text{m}$  and 9-12  $\mu\text{m}$  part of the spectrum, respectively. The lasers can be constructed so that they will oscillate on one line at a time, with stability and power levels that are appropriate for use in a LAS.

Since carbon monoxide and carbon dioxide lasers are only discreetly tunable, it has been necessary to investigate the spectral overlaps that exist between their emission lines and the absorption lines of the interesting pollutants. A significant amount of laboratory work has revealed that many such overlaps do occur (Refs. 5, 6, 7). Table 1 presents a summary of some of the important atmospheric pollutants that have absorption lines coincident with CO and CO<sub>2</sub> laser emission lines.

**Table 1**

Pollutant	Absorption Band Center	Laser Line
Nitric Oxide NO	5.3- $\mu\text{m}$	CO: 7-6 band, P(13) 7-6 band, P(15) 9-8 band, P(9) 9-8 band, P(13) CO <sub>2</sub> (doubled): 00 <sup>0</sup> 1-10 <sup>0</sup> 0 band, P(8) P(24) R(8) R(10)
NO <sub>2</sub>	6.2- $\mu\text{m}$	CO: 18-17 band, P(13) 19-18 band, P(8) 20-19 band, P(10) 22-21 band, P(10)
SO <sub>2</sub>	7.4- $\mu\text{m}$	CO: 31-30 band, P(9) 32-31 band, P(6)
SO <sub>2</sub>	8.7- $\mu\text{m}$	C <sup>12</sup> O <sub>2</sub> <sup>18</sup> : 00 <sup>0</sup> 1-10 <sup>0</sup> 0 band, R(42) R(44)
CO	4.7- $\mu\text{m}$	CO <sub>2</sub> (doubled): 00 <sup>0</sup> 1-02 <sup>0</sup> 0 band, P(24) R(18)
O <sub>3</sub>	9.5- $\mu\text{m}$	CO <sub>2</sub> : 00 <sup>0</sup> 1-02 <sup>0</sup> 0 band, P(14)
NH <sub>3</sub>	10.5- $\mu\text{m}$	CO <sub>2</sub> : 00 <sup>0</sup> 1-10 <sup>0</sup> 0 band, P(32)

A typical absorption line of a trace gas in an atmospheric background has a linewidth of  $0.1 \text{ cm}^{-1}$ . Expressed in frequency units, this linewidth is about 3 GHz. Laser linewidths are significantly smaller, usually less than 100 KHz in a well designed and passively stabilized laser. Furthermore, these lasers can be tuned within the pressure broadened width of the spectral line being used.. Our present laboratory CO and CO<sub>2</sub> lasers (Ref. 8) operate at pressures in the 10-20 torr range, and can be tuned  $\pm 15$  MHz from line center. The newly developed waveguide molecular lasers operate at pressures in the 100-300 torr range, and can be tuned over  $\pm 1$  GHz from the line center (Refs. 9, 10). The ability to tune the laser source over a good portion of an absorption line of a pollutant molecule is an important advantage, and will allow us to learn much about the spectra of molecules that has not been attainable with more conventional spectroscopic techniques.

In order to calculate the laser power required to perform a measurement of absorption loss, we must compare the intensity of the return signal from the ground and the heterodyne receiver's inherent internal noise. The amount of transmitted power,  $P_t$ , collected by the receiving aperture, diameter  $d$ , in an aircraft at altitude  $h$  is

$$P_r = rP_t \frac{d^2}{4h^2} \quad (1)$$

where  $r$  includes all the losses along the path. The sensitivity of the heterodyne receiver is best expressed in terms of the noise-equivalent-power (NEP) of an optical heterodyne radiometer, since the receiving system cannot be considered to be coherent with the transmitted signal due to the ground surface roughness. The NEP is (see Ref. 1):

$$\text{NEP} = \frac{2h}{\eta} \sqrt{\frac{B_{if}}{\tau}} \quad (2)$$

where  $\eta$  is the detector quantum efficiency,  $B_{if}$  is the bandwidth of intermediate frequency amplifier, and  $\tau$  is the post detection integration time. If the system is operating at a wavelength near  $10 \text{ }\mu\text{m}$ , then with  $\eta = 50\%$ ,  $B_{if} = 1 \text{ MHz}$ , and  $\tau = 0.1 \text{ sec}$ , the NEP is  $2.4 \times 10^{-16}$  watts. For operation from an altitude of 10 km, a 1-watt transmitter and a collecting aperture of 5 cm diameter would provide a signal-to-noise ratio of about 2000.

It should be noted that the main source of noise in this type of optical receiver is the internal noise of the detector itself. The bandwidth of the receiver is very narrow by optical standards, and the noise amplitudes of both the Earth's thermal emissions and the scattered sunlight are quite small by comparison. The system is thus free of the interfering effects that are a source of difficulty to many infrared systems.

In order to detect a pollutant, it is necessary to measure the path loss at one of the overlapping wavelengths mentioned above. Operation at a second, nearby wavelength is

used to calibrate the fixed path losses, with the difference in the two measurements being attributed to the absorption by the pollutant. The main limitation to detection of small levels of a pollutant is the ability to accurately measure the difference between the two signals. For an instrument mounted in an aircraft, measurements must be made simultaneously at two wavelengths, since the Earth surface albedo will fluctuate very rapidly. Assuming that the minimum signal difference that we can observe is 1%, and using previously published data for absorption losses at laser wavelengths we can calculate the amount of pollutant necessary to produce a measurable absorption loss. Table 2 presents a summary of minimum detectable levels of common atmospheric pollutants. Also presented in Table 2 are comments about the pollutant levels to be expected on a typical smoggy day. Many other materials that are components of photochemical smog can also be detected by laser absorption techniques. More extensive laboratory study of these materials is necessary, however, before we can consider monitoring them remotely. Among these materials are carbon monoxide, peroxyacetyl nitrate, aldehydes and other hydrocarbons.

**Description.** The details of one version of a laser absorption spectrometer will now be presented. The instrument is to be mounted in an aircraft, and will be used to monitor ozone concentrations and flow patterns in the South Coast Air Basin of California. With minor modifications, the instrument can be used to monitor other atmospheric constituents, with particular emphasis being placed upon the oxides of nitrogen.

The aircraft instrument consists of two identical laser transmitter-heterodyne receiver systems operating simultaneously along the same atmospheric path. The two lasers will be operating at different wavelengths, and the measured parameter is the differential absorption. At closely spaced wavelengths, the differential effects of the albedo fluctuations, as well as atmospheric turbulence, aerosol scattering and optical system losses will be minimal, and the main observable difference will be difference in absorption loss due to the molecular species along the atmospheric path.

Figure 2 presents a schematic diagram of the optical portion of the instrument. The lasers chosen for the first series of experiments are waveguide type carbon dioxide lasers. They are constructed so that individual lines can be selected with a Littrow-mounted diffraction grating serving as one mirror of the laser, and with the other mirror mounted on a piezoelectric translator to provide fine tuning of the laser's frequency. The laser is designed so that it will not require active stabilization for its operation. These lasers are capable of one watt continuous output at most of the CO<sub>2</sub> lines of both the 9.4 μm and 10.4 μm bands. The laser signals are optically chopped to provide suitable modulation for the power monitor and receiver electronics. The beams will be transmitted through small telescopes that will aid in controlling the beam divergence and assure proper boresighting

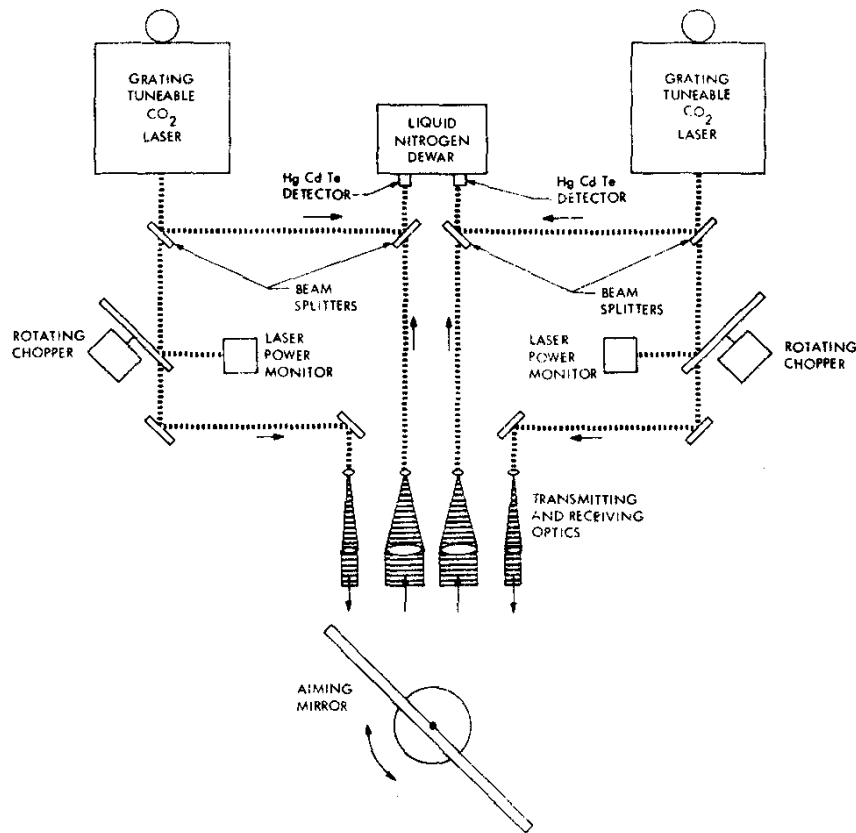
**Table 2. Sensitivities to Various Pollutants**

Pollutant	Amount Necessary for a 1% Absorption (ppb-km)	Laser Line	Spectral Region	Comments
Ozone	2.5	$C^{12}O_2^{16}$ laser: 9.4- $\mu$ m band, P(14)	9.5- $\mu$ m	Normal ground level concentrations are about 10 ppb; reaches the several hundred ppb level in photochemical smog; stratospheric ozone peaks around 25 km, where the mixing ratio is about 8 ppm.
Ethylene	1.5	$C^{12}O_2^{16}$ laser: 10.4- $\mu$ m band, P(14)	10.5- $\mu$ m	A man-made constituent; negligible amount in clean air; concentrations as high as 1 ppm in high vehicle density regions.
Ammonia	2.5	$C^{12}O_2^{16}$ laser: 10.4- $\mu$ m band, P(32)	10.5- $\mu$ m	Normal concentrations are 5-20 ppb; originates in soil; interacts with $SO_2$ and participates in particulate formation.
Sulphur Dioxide	90.0	$C^{12}O_2^{18}$ laser: 9.2- $\mu$ m band, R(40)	9.0- $\mu$ m	Normally around 10 ppb; reaches concentrations of several hundred ppb in polluted environments.
Nitric Oxide	15.0	CO laser: 7-6 band, P(15)	5.2- $\mu$ m	Normally concentrations are a few ppb, due to soil bacterial action; concentrations reach the several hundred ppb level in polluted urban areas, due to combustion processes.
Nitrogen Dioxide	20.0	CO laser: 18-17 band, P(13)	6.0- $\mu$ m	Normally a few ppb; concentrations reach the several hundred ppb level in polluted areas, due to oxidation of NO.

Assembly for the Aircraft Laser Absorption Spectrometer of the two beams. A small portion of each laser signal is split off to provide local oscillator signals for the receiver system.

In order for proper operation of the heterodyne receivers, a frequency shift between the local oscillator signal and the return signal is necessary. A very simple method for providing this frequency shift is to take advantage of the aircraft's velocity to produce a Doppler shift in the signal scattering from the ground. By aiming the system field-of-view via a steerable mirror forward along the aircraft's ground track, adequate frequency shifts for the receivers can be obtained.

The receivers are identical, consisting of 5 cm diameter beam collecting telescopes and liquid nitrogen cooled mercury-cadmium-telluride detectors. Two signals are projected onto each detector: the local oscillator signal and the return signal from the ground. The detectors have sufficiently short time constants that the signal at the difference frequency, the intermediate frequency, is readily available. Note that this photomixing process is inherent to any optical detector (Ref. 11), since these detectors respond linearly to optical power. This process does not depend upon an intrinsic optical nonlinearity of the device.

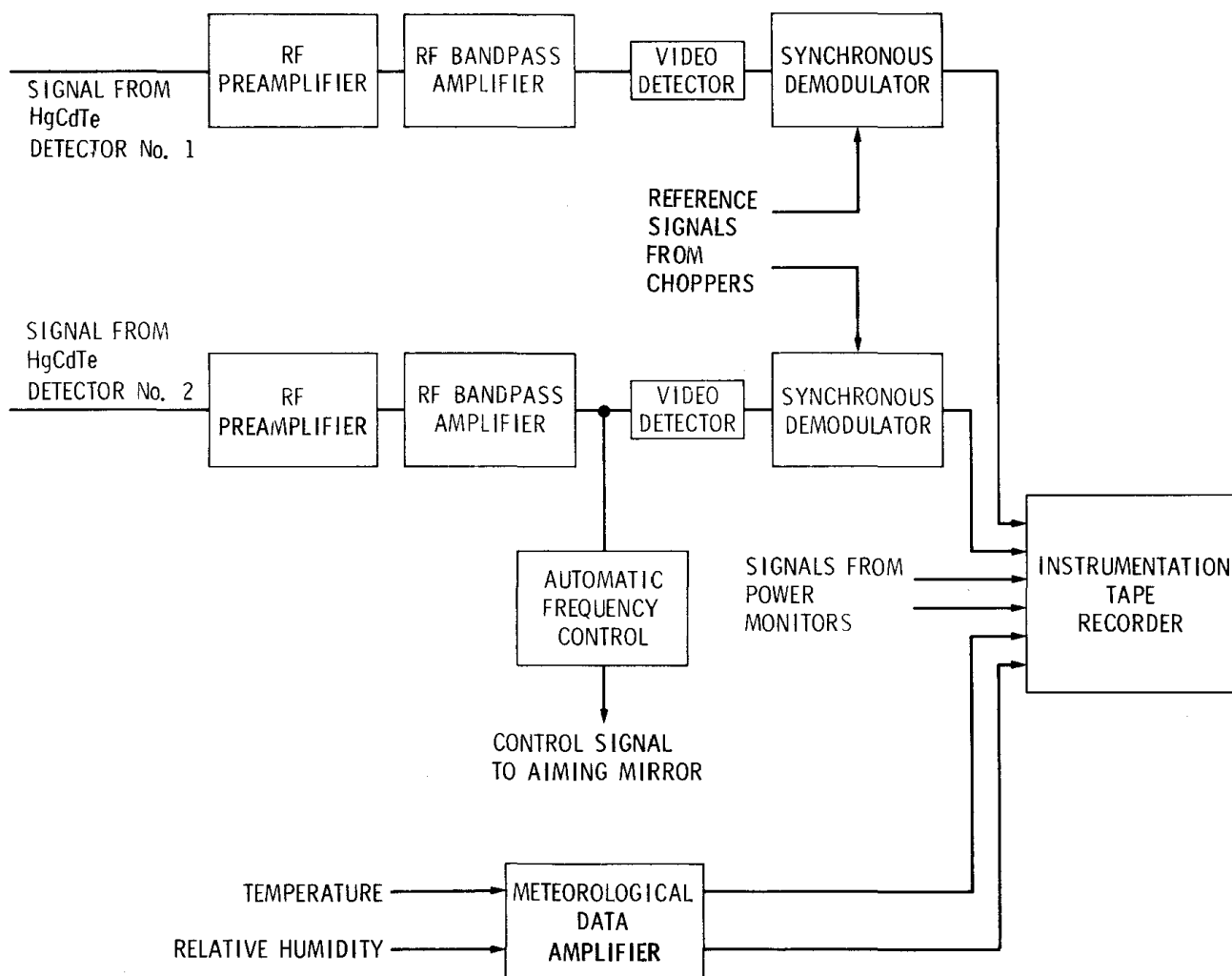


**Figure 2. Schematic Diagram of the Transmitter-Receiver Assembly for the Aircraft Laser Absorption Spectrometer**

The block diagram of the receiver electronics and other ancillary equipment is presented in Figure 3. The signal path electronics for each receiver is identical, consisting of an RF preamplifier, a bandpass amplifier, a video-type detector, and a synchronous demodulator and filtering amplifier. The signals from each receiver, along with laser power monitor signals and necessary meteorological data are all recorded on an instrumentation-type tape recorder or telemetered directly to a central ground station.

In order to provide adequate control of the Doppler-produced frequency difference, an automatic frequency control is incorporated into the system. The AFC adjusts the angle of the aiming mirror to keep the difference frequency within the bandpass of the receiver electronics.

In principal, the reduction of the recorded data to produce a record of the pollutant concentration is relatively simple. The recorded data can be played back into either a special purpose analog system, or digitized and processed by a digital computer. Either way, the calculations required are trivial, and little time would be required. If a real time recording is deemed necessary, additional equipment in the aircraft would provide for it. The reduction of the data could be complicated by the presence of an atmospheric



**Figure 3. Block Diagram of the Receiver Electronics and Recording Equipment.**

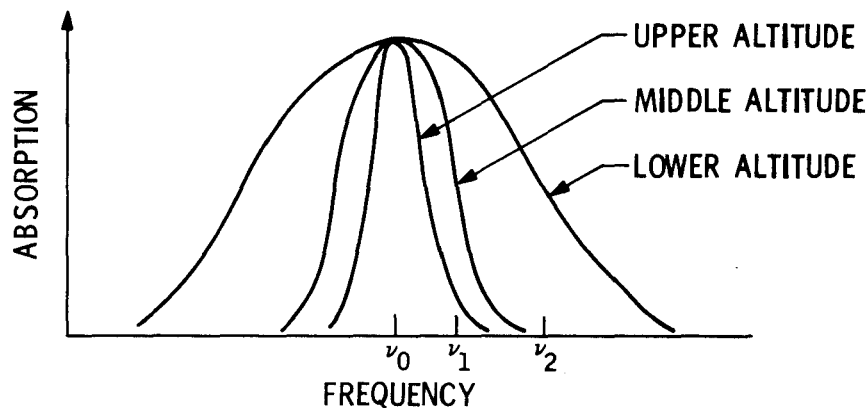
constituent whose absorption lines overlapped the two wavelengths, chosen to operate the LAS. A way of overcoming this problem is to operate at additional wavelengths, requiring additional transmitter-receiver systems. The data reduction then becomes quite involved, requiring the use of a digital computer to perform the solution to a complex matrix equation. Such interference effects are expected to be minimal with most atmospheric constituents.

**Applications.** The most significant application of the laser absorption spectrometer is remote monitoring of selected pollutants over a wide area. The instrument has the capability of performing an additional measurement that is of great interest to many atmospheric physicists. The waveguide lasers being used are able to tune over the considerable portion of an operating line, a frequency range of about a GHz. By tuning the laser to several different points on an atmospheric absorption line, the line shape can be deduced. Detailed knowledge of an absorption line shape, as measured over a long vertical



path through the atmosphere, is sufficient to allow calculation of the variation of concentration with altitude of the pollutant being monitored.

The technique for determining the altitude profile of a pollutant from its absorption line shape has been extensively studied theoretically and is reported in Reference 12. The technique utilizes the fact that the absorption linewidth varies with altitude, due to the pressure variations. Absorption lines are narrower at higher altitudes, and broader at lower altitudes. Higher altitude pollutants contribute more to the total absorption near the center of a line than they do at frequencies that are displaced from the line center. Figure 4 illustrates this effect. A total absorption measurement at the line center,  $\nu_0$ , would determine the contributions from all altitudes, and would be used to determine the total pollutant burden. Measurement at  $\nu_1$ , would show the contribution from the middle and lower altitude pollutants, and measurement at  $\nu_2$ , the lower pollutants. Computer simulations of several interesting pollutants and altitude profiles have been performed. The results indicate that the resolution of the profile measurement is 4 to 5 km in upper regions of the atmosphere and 2 to 3 km in the lower atmosphere. This resolution is adequate to determine inversion layer altitudes, and would be of significant aid in monitoring the effects of wind on mixing and distributing pollutant loads.



**Figure 4.** The altitude profile principle is based on the fact that absorption line widths are proportional to background pressure and only slightly dependent on temperature. Measurement of total absorption at several frequencies (e.g.  $\nu_0$ ,  $\nu_1$ ,  $\nu_2$ ) provide concentrations at various altitudes.

The aircraft-borne laser absorption spectrometer as it is presently designed will be of great utility in studying photochemical smog. It will have the capability to determine the spatial distribution of selected pollutants over reasonably large geographic areas. An improved 'Version of this instrument can be considered when the tunable laser technology has advanced to the point where high power, broadly tunable (but narrow linewidth) lasers become available. An instrument based upon the same concept will function from typical Earth satellite altitudes. Such an instrument would provide an opportunity to monitor trace

atmospheric constituents on a global basis, and this would be a very useful tool to aid man in his efforts to assess the effects of his impact on his environment.

## REFERENCES

1. Menzies, R. T., and Shumate, M.S. "Usefulness of the Infrared Heterodyne Radiometer in the Remote Sensing of Atmospheric Pollutants, AIAA Paper No. 71-1083 presented at the Joint Conference on Sensing Environmental Pollutants, Palo Alto, CA., November 8-10, 1971.
2. Hinkley, E. D., and Kelley, P. L., *Science* 171, 635 (1971).
3. Nill, K.W., Strauss, A. J., and Blum, F.A., *Appl. Phys. Lett.* 22, 677, 15 June 1973.
4. Hinkley, E.D., "Bistatic Monitoring of Gaseous Pollutants with Tunable Semiconductor Lasers", 1974 Pittsburgh Conf. on Analytical Chemistry and Applied Spectroscopy, Cleveland, Ohio, 7 March 1974.
5. Menzies, R. T., George, N., and Bhaumik, M. L., *I.E.E.E. J. Quantum Electron.* 6, 800 (1970).
6. Menzies, R. T., *Appl. Phys. Lett* 22, 592 (1 June 73).
7. Menzies, R. T., and Shumate, M.S., *Science* 184, 570 (1974).
8. Freed, C., *Appl. Phys. Lett.* 18, 458 (15 May 1971).
9. Abrams, R. L., and Bridges, William B., *I.E.E.E. J. Quantum Electron.* QE-9, 940 (September 1973).
10. Asawa, C.N., *Appl. Phys. Lett.* 24, 121 (1974).
11. Ross, M., "Laser Receivers", p. 109, Wiley, New York (1966).
12. Menzies, R. T. and Chahine, M. T., "Remote Atmospheric Sensing with an Airborne Laser Absorption Spectrometer," to be published