ATMOSPHERIC MONITORING USING INFRARED HETERODYNE RADIOMETRY

BERNARD J. PEYTON
AIL
Cutier-Hammer
Melville, N.Y.

Summary. Atmospheric constituents have unique vibrational-rotational signature lines within the infrared spectrum and the signature intensities and line shapes vary with the constituent concentration and the atmospheric density. The recent development of stable, single frequency, single-mode laser local oscillators and nearly quantum-noise-limited heterodyne receivers have permitted the development of infrared heterodyne radiometers (IHR's) which provide good sensitivity and excellent specificity for the remote examination of individual atmospheric constituent signature lines. A 9 to 11 µm IHR employing a CO_2 laser local oscillator has been developed and can be used to resolve the spectral signature of atmospheric constituents such as SO_2, O_3 C_2H_4, and NH_3. The IHR has a bandwidth of 100 MHz (33×10^{-3} cm^{-1}) and a minimum detectable power spectral density of 5.4×10^{-24} W/Hz for a 1-second integration time. For atmospheric monitoring applications the IHR telescope collects the thermal energy radiating from the earth at: (1) a clear spectral window, and (2) a spectral region in which the signature lines of the constituent gases at various layers of the atmosphere will be energized by the upwelling thermal radiation. When the vertical temperature distribution of the atmosphere is known, the concentration of the atmospheric constituent gas can be determined as a function of altitude from the radiance data collected at the IHR using an iterative mathematical technique.

Introduction. Remote measurements of the concentration and vertical distribution of atmospheric constituents on a global and regional basis have been carried out at infrared wavelengths using direct detection techniques (ref 1 and 2). For example, the Infrared Interferometer Spectrometer (IRIS) operates between 400 and 1600 cm^{-1} with a spectral resolution of 84 GHz (2.8 cm^{-1}) and a noise equivalent radiance (NEP) of 0.5 erg s^{-1} cm^{-2} sr^{-1}/cm^{-1} (ref 1). Since the infrared signature line widths of atmospheric constituents are approximately 3 GHz (10^{-1} cm^{-1} near ground level and decrease to Doppler limited linewidths near 100 MHz (3.3×10^{-3} cm^{-1}) at high altitude, the direct detection type of infrared instrumentation will examine a portion of the infrared spectrum which includes a significant number of constituent signature lines. In addition, the direct detection instrumentation may collect thermal radiation which is strongly overlapped by thermal

emission lines of atmospheric gases other than the constituents being monitored. This paper considers the potential of infrared heterodyne radiometers (IHR's) for the remote passive monitoring of atmospheric constituents. The infrared heterodyne receiver provides excellent sensitivity and specificity when compared to conventional infrared detectors (ref 3) and can be tuned to discrete portions of the infrared spectrum by proper selection of the laser local oscillator (LO).

An IHR has been developed (ref 4) for operation at the CO_2 laser wavelengths (9 to 11 µm) which exhibits an IF bandwidth between 2 MHz (7×10^{-5} cm^{-1}) and 100 MHz (3.3×10^{-3} cm^{-1}) and a minimum detectable power spectral density of 5.4×10^{-24} W/Hz for a 1-second integration time. The 9 to 11 µm IHR was used to carry out atmospheric propagation measurements (ref 4). A wideband 9 to 11 gm IHR can be used to resolve the spectral signature of atmospheric constituents such as SO_2, O_3, C_2H_4, and NH_3 by measuring the upwelling thermal radiation at specific infrared signature lines (Table I).

Table I. Pollutants Having Infrared Signatures Which Overlap
CO_2 Laser Transitions

Pollutant	Source	Location (µm)	Laser Line (µm)
O_3 (Ozone)	Atmospheric photo-chemistry	9.6	CO_2--9.6
C_2H_4 (Ethylene)	Engine exhaust	10.5	CO_2-- 10.53
NH_3 (Ammonia)	Organic waste	10.5	CO_2-- 10.6
SO_2 (Sulfur Dioxide)	Burning of fuels	9.018	CO_2--9.018 (isotope doped)
CH_3OH (Methanol)	Engine exhaust	9.67	CO_2--9.67

Infrared Heterodyne Radiometers. The Dicke-type infrared heterodyne radiometer consists of a collecting telescope, a Dicke switch, an infrared photomixer, a laser local oscillator, an IF amplifier chain, and a radiometric processing network. Heterodyne detection is most practical at the longer infrared wavelengths due to reduced photon noise (hv), easier alignment of source energy and laser local oscillator, and the larger diffraction limited field of view.

The remote sensing of selected optical line signatures can be best achieved with an IHR by: (1) using a tunable receiver to detect spectral lines which fall within an atmospheric transmission window, (2) matching the spectral window of the receiver to the selected emission (or absorption) signature line, (3) providing a real-time reference by rapidly

switching between the selected spectral line and a nearby "empty" wavelength band, (4) using integration techniques to sum the spectral signature over a large number of measurement intervals, and (5) using an optical system in which a small instantaneous field of view is scanned to cover a large spatial region.

The instantaneous IF bandwidth of the receiver establishes the frequency (spectral) resolution of the IHR and its lower limit is fixed by laser instabilities (<1 MHz), such that the Doppler widths of tens of MHz can be resolved. Thus, an IHR with an instantaneous bandwidth of 1.5 GHz (an overall spectral window of 3 GHz) and a narrow tunable IF window will permit the spectral inspection of a molecular signature on a real-time basis. The conventional CO_2 laser is tunable over a 40-MHz band; additional tuning ($\pm$ 700 MHz) is achievable in waveguide lasers using pressure broadening techniques. Fig. 1 illustrates that infrared (LO) tuning is a more effective way of defining the signature profile than electronic (IF) tuning since proper tuning of the laser LO will include the infrared image of the photomixer within the infrared line spectra. Cooled diode lasers which exhibit wide tunability and low available power may be potential laser LO sources.

An infrared heterodyne receiver can be designed to provide nearly quantum-noise-limited sensitivity by fixing the mean-square shot noise current induced by the absorbed laser LO power to be significantly larger than the sum of all other receiver noise currents. For a reversed biased photovoltaic photomixer, the heterodyne receiver sensitivity is given (ref 3) by

$$\frac{NEP(f)}{B} = \frac{h\nu}{\eta} \left\{ 1 + \frac{2k\,(T_m + T'_{IF})}{q\,I_o}\,G_D\,\left[1 + (f/f_c)^2\right] \right\} \tag{1}$$

where NEP is the noise equivalent power for a unity receiver signal-tonoise ratio, h is Planck's constant, ν is the infrared frequency, η is the photomixer quantum efficiency, B is the IF bandwidth, k is Boltzmann's constant, T_m is the photomixer temperature, T_{IF} is the effective input noise temperature of the IF amplifier, q is the electronic charge, I_o is the laser LO induced dc photocurrent, G_D is the small signal shunt conductance, f is the IF frequency, and f_c is the 3-dB cutoff frequency of the photomixer. Equation 1 can be simplified to

$$NEP = \frac{h\nu B}{\eta'} \tag{2}$$

where $\acute{\eta}$ is the effective receiver quantum efficiency. When observing a thermal source the IHR predetection signal-to-noise ratio is given by

$$\left(\frac{S}{N}\right)_p = \frac{h\nu B\,T_o}{e^{(h\nu/kT_s)} - 1} \cdot \frac{\eta'}{h\nu B} = \frac{\eta'\,T_o}{e^{(h\nu/kT_s)} - 1} \tag{3}$$

where τ is the IHR integration time and T_o is the transmissivity between the thermal source and the IHR.

When measuring atmospheric constituents, an IHR with a wideband photomixer/preamplifier combination should be employed to examine the collision broadened signature line. For remote sensing applications the photomixer should have a 3-dB cutoff frequency near 1 GHz, operate at a temperature near 100 K, require minimum laser LO and dc bias power, and be suitable for aircraft or spacecraft operation. Previously developed PV:HgCdTe photomixers mounted in a specially designed housing (Fig. 2) have been shown to meet these technical objectives. Cutoff frequency (Fig. 3) and RFI measurements on the PV:HgCdTe photomixers indicate that the detector housing could be effectively used above 1000 MHz. Laboratory measurements on a heterodyne receiver having a selected infrared mixer, an optimized impedance matching network, and a specially designed wideband IF preamplifier indicate that an infrared heterodyne receiver having an NEP <1.5 x 10^{-19} W/Hz at 1000 MHz and ≤ 2.0 x 10^{-19} W/Hz at 1500 MHz at CO_2 laser wavelengths is presently achievable for a photomixer temperature of 125 K and incident laser LO power levels near 2 mW.

The temperature resolution of a previously developed IHR (Fig. 4) has been measured using a blackbody radiator as the source. The blackbody thermal radiator was positioned in the field of view of the radiometer telescope and a simple lens was used to collimate the thermal source. For the experimental conditions of $\lambda = 10.6$ µm, B = 100 MHz, $\tau = 3$ s, and NEP = 2 x 10^{-19} W/Hz, the IHR output was monitored as the thermal source temperature was varied. The calculated and measured IHR temperature resolution of the IHR is given in Fig. 5. The measured temperature resolution of the IHR was approximately $\Delta T_S = 15$ K for a source temperature of $T_s = 300$ K and decreased to a $\Delta T_s = 3$ K for a source temperature of $T_s = 1000$ K. The IHR temperature resolution is expected to be reduced to approximately 0.8 K at $T_s = 300$ K for an IF bandwidth of 1500 MHz.

The slight variation between the calculated and measured resolution is attributed to: (1) optics losses, (2) a slight astigmatism in the off-aids parabolic telescope, and (3) some rapid amplitude fluctuations in the CO_2 laser LO. The use of reflective optics, higher quantum efficiency photomixers, and a 1500-MHz IF bandwidth may reduce the IHR temperature resolution below 0.5 K for $T_s = 300$ K. It should also be noted that the IHR temperature resolution can also be improved by increasing the system integration time.

Two possible spatial measurement modes exist for the remote sensing of atmospheric constituents: (1) a quasi-fixed platform such as a stationary satellite (or balloon platform) can be used in conjunction with a scanning optical system., or (2) a moving platform such as a nearearth spacecraft (or aircraft) can be used to continuously scan portions of the earth and its environment. The telescope requirements will be fixed by the particular

vehicle and mission characteristics. For the case of an IHR on a synchronous satellite, a 10-inch diameter telescope having a diffraction-limited half beam width of 130 µrad will resolve a 5-km diameter area on earth. The use of infrared heterodyne radiometers for remote sensing applications has been previously discussed by several authors (ref 5 to 11) . IHR applications such as solar physics, astronomy, atmospheric physics, and plume physics have been considered (ref 4 to 11).

Vertical Profiling Using an IHR.   For vertical profiling applications, the laser local oscillator of the IHR is tuned to overlap a selected signature line of an atmospheric constituent and the optical telescope collects the upwelling thermal energy radiating from the earth. The thermal radiation energizes gas molecules located at the various layers of the atmosphere and variations in the incident thermal radiation at any atmospheric layer (Fig. 6) causes changes in the quantum-mechanical population distribution of the energy states of the gases. For a selected constituent the energy state differences correspond to unique "signature" frequencies given by

$$f_N = \frac{\Delta E_N}{h} \tag{4}$$

where ΔE_N is the energy state difference between the two levels of the selected atmospheric constituent. The transition of particles between the energy states of an atmospheric gas results in an absorption (emission) at the infrared signature frequency. The absorption (emission) strength at the signature frequency is given by

$$N_i = AN\, e^{-E_i hf/kT} \tag{5}$$

where N_i is the number of gas particles in the ith state, A is a gas constant, N is the total number of constituent particles, E is the energy constant of the ith state, k is Boltzmann's constant, and T is the gas temperature.

The vibrational and rotational motion of atoms in the molecule of the selected atmospheric constituent result in an infrared absorption (emission) spectra. Infrared wavelengths which are larger than 3.5 µm are particularly suited for passive atmospheric monitoring applications because the thermal emission from the earth at these wavelengths is greater than the energy due to reflected and scattered sunlight. The location of signature lines due to mid-infrared vibrational-rotational transitions are an intrinsic characteristic of the selected atmospheric constituent. The intensity of a selected signature line is a characteristic of the gas constituent, the gas temperature, and the energy state being monitored. The spectral line width of a signature line is characteristic of the temperature and/or the pressure as well as the specific weight of the selected constituent. For example, the measured variation of the spectral line width of Ammonia (NH_3) with pressure is given in Fig. 7. The measured Doppler line width for NH_3 is 100 MHz and the signature

line width increases to approximately 1 GHz near P = 5 torr due to collision broadening. Measured laboratory data on the variation of pollutant line width with pressure can be used to determine vertical distribution of pollutants by IF frequency examination of the total spectral profile collected at the IHR.

For atmospheric constituents with known vertical concentration distributions, remote IHR measurements can be used to establish temperature profiles of the atmosphere for meterological applications. On the other hand, for the condition of a known temperature profile of the atmosphere normally obtained from available data, remote IHR measurements can be used to establish the vertical concentration profile of an atmospheric constituent. The basic equation for the radiance collected at an IHR receiver which: (1) looks at the earth, (2) is located at an altitude $y = \ell$ (Fig. 6), and (3) spectrally overlaps the atmospheric constituent signature line of interest, is given by

$$N_i = B_o \tau_o + \sum_{o}^{y=\ell} B_i \frac{\partial \tau_i}{\partial H} \Delta H \tag{6}$$

where $B_i(y) = f(T)$ is the thermal irradiance over the spectral width of the IHR receiver at altitude y, B_o is the irradiance at the surface of the earth (y = o) due to the earth's ambient temperature, $\tau_i(y)$ is the average spectral transmission of the atmosphere from altitude $y = y_i$ to altitude $y = \ell$ given as a function of the spectral signature characteristics of the absorbing atmospheric gas, and τ_o is the average spectral transmission from the earth (y = o) to the IHR (y = ℓ). The first term on the right in equation 6 is the net radiance due to the earth emission as attenuated by the absorbing atmospheric gas in the path between the earth (y = o) and the IHR (y = ℓ). The second term of equation 6 represents the reemission radiances at each level y_i, subject to attenuation in the atmospheric path between levels $y = y_i$ and $y = \ell$.

As may be seen from equation 6, the two basic quantities which enter into the radiance equation are the atmospheric irradiance and atmospheric transmission. The atmospheric irradiance is temperature dependent and the atmospheric transmission is dependent on the spectral signature and the concentration of the pollutant gas. Thus a knowledge of the atmospheric profile of either one of the basic quantities is required in order to determine the other.

From Planck's radiation equation, the thermal irradiance of a source at temperature T is given by:

$$W\lambda = \frac{2\pi hc^2}{\lambda^5} \cdot \frac{1}{e^{ch/\lambda kT} - 1} \left(\frac{W}{cm^2 \, cm^{-1}} \right) \tag{7}$$

or

$$W_f = \frac{2\pi\,hc^2}{\lambda^3} \cdot \frac{1}{e^{ch/\lambda kT} - 1} \left(\frac{W}{cm^2\,Hz}\right) \tag{8}$$

where c is the speed of light. The radiance is transmitted through a uniform atmosphere with a transmission coefficient which is given by:

$$\tau_i = e^{-\int_{H_i}^{\ell} \alpha_i\,W_i\,\frac{P(H_i)}{P_o}\,d\,H} \tag{9}$$

and the line absorption coefficient for a collision broadened line is given by:

$$\alpha_i = \frac{S}{\pi}\,\frac{\gamma}{(f - f_o)^2 + \gamma^2} \tag{10}$$

where $P(H_i)$ is the atmospheric pressure at $y = H_i$, P_o is the atmospheric pressure at sea level (y o), W_i is the atmospheric constituent concentration at the ith level, ℓ is the total atmospheric path, S is the absorption (emission) line intensity, f_o is the center frequency of the absorption line, and γ is the half-width at half-maximum of the absorption line.

The constituent concentration as a function of altitude is determined using the radiance data collected at the IHR. For a measured radiance N_i an inversion of the basic relationship (equation 6) is required so as to solve for the constituent concentration W_i as a function of $\tau_i(y)$ for the case of $\tau_i(y)$ being a function of N_i. Since no unique explicit solution of the localized constituent concentration W_i in terms of the measurable quantity N_i is possible, the data reduction is carried out using an iterative solution technique (ref 11 to 13) . The key to the iterative process is based on derived relationships between the actual pollutant concentration, which is reflected in the radiance measured at the MR, and an assumed pollutant concentration, which deviates by a small value from the actual measured radiance. The ratio of the pollutant concentrations is proportional to a function of the linear difference between the radiances. Thus by a consecutive iteration, the difference is made small until the actual concentration is approached.

The accuracy of the process may be enhanced by employing several channels and weighing the obtained parameters over the outputs of each channel. The maximum information at a given altitude level is obtained from a channel which has a maximum slope of its transmission function at the given level (ref 11) . Hence, a judicious selection of the spectral channels (or laser local oscillators) appears to be important for optimum accuracy and/or vertical resolution.

Conclusions. The use of infrared heterodyne radiometers to remotely monitor the concentration and vertical distribution of selected atmospheric constituents has been reviewed. A 9 to 11 µm IHR has been developed for atmospheric propagation measurements and appears to be uniquely suitable for atmospheric monitoring applications. A 9 to 11 µm IHR having an IF bandwidth of 1.5 GHz is presently being developed for use in an aircraft (or balloon) for the remote measurements of atmospheric constituents such as SO_2, O_3, C_2H_4, and NH_3.

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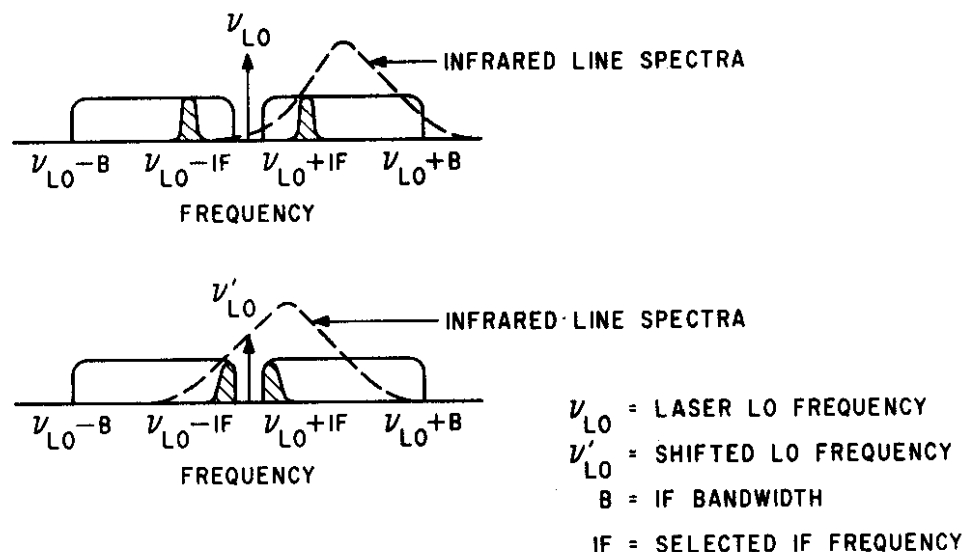
FIGURE 1.
USE OF ELECTRONIC AND INFRARED TUNI NG OF IHR SPECTRAL
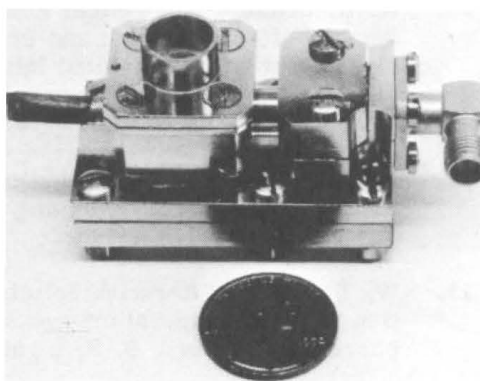RESPONSE TO EXAMINE INFRARED SIGNATURE LINE

FIGURE 2.
SPACE-QUALITY PHOTOMIXER AND HOUSING SUITABLE
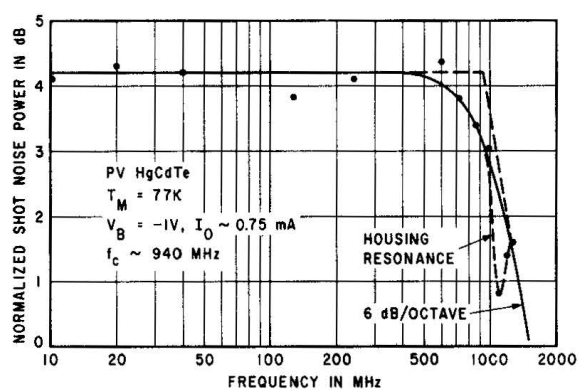FOR USE AT CO_2 LASER WAVELENGTH

FIGURE 3.
MEASURED FREQUENCY RESPONSE OF PV:HgCdTe PHOTOMIXER
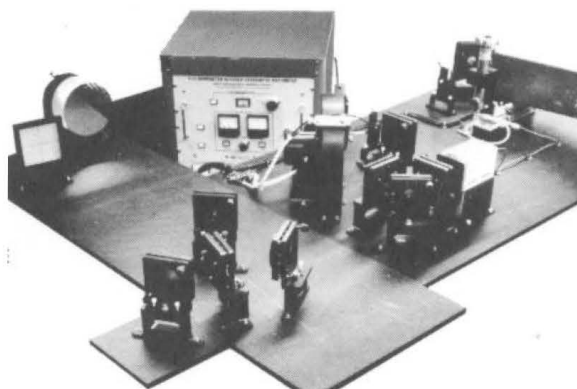MOUNTED IN SPACE-QUALITY HOUSING

FIGURE 4.
9 TO 11 μm INFRARED HETERODYNE RADIOMETER

30.0

λ = 10.6 μm
B = 100 MHz
$\tau_{(RC)}$ = 3 SECONDS
NEP $\approx$ 2 X 10^{-19} W/Hz

TEMPERATURE RESOLUTION ΔT IN K
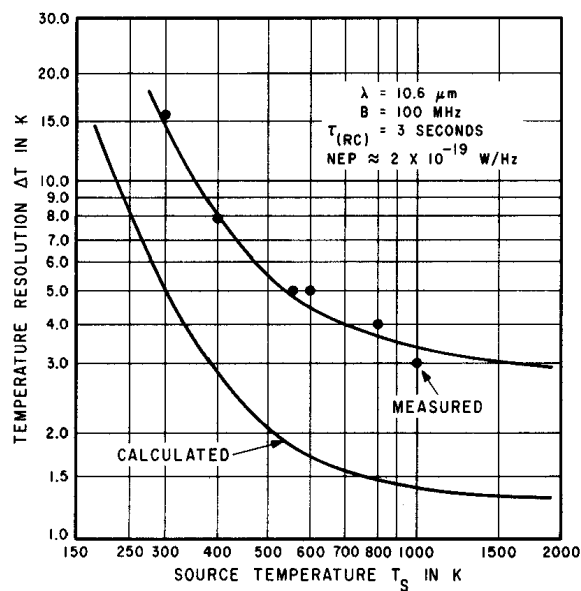
MEASURED

CALCULATED

SOURCE TEMPERATURE T_S IN K

FIGURE 5.
CALCULATED AND MEASURED
TEMPERATURE RESOLUTION
OF IHR AS A FUNCTION OF
SOURCE TEMPERATURE

FIGURE 6.
SIMPLIFIED REPRESENTATION OF THE
USE OF AN INFRARED HETERODYNE
RADIOMETER TO MONITOR
ATMOSPHERIC CONSTITUENTS

T_n = ATMOSPHERIC CONSTITUENT TEMPERATURE

W_n = ATMOSPHERIC CONSTITUENT CONCENTRATION

$\lambda \sim$ 10.5 μm
T = 300 K
L = 10 cm
δB = 10^5 Hz
$\Delta f_d \sim$ 85 MHz

RELATIVE TRANSMISSION

100 MHz

0.5 TORR

0.005 TORR

0.05 TORR

5 TORR
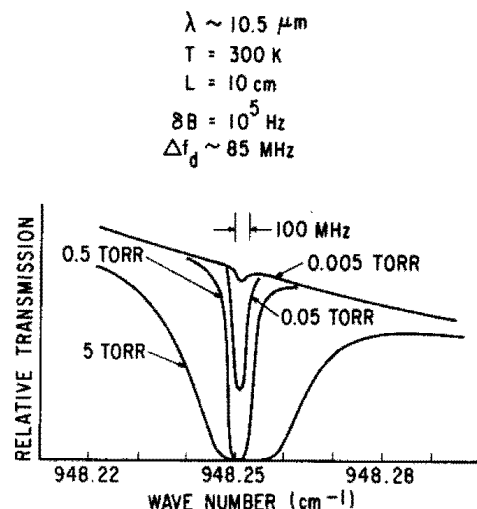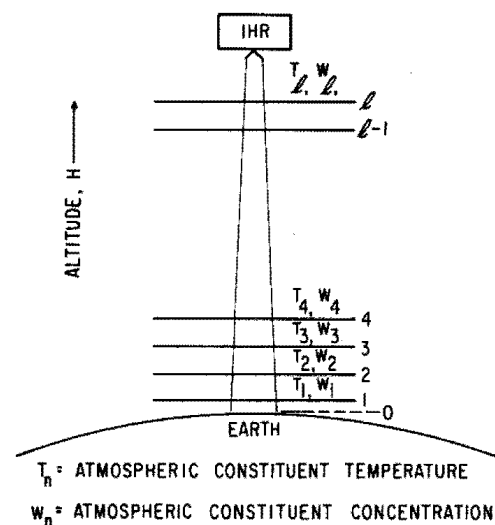
948.22   948.25   948.28

WAVE NUMBER (cm^{-1})

FIGURE 7.
MEASURED ABSORPTION SPECTRA
OF sP (1, 0) LINE OF AMMONIA
MOLECULAR POLLUTANT IN
LABORATORY ENVIRONMENT
(AFTER HINKLY AND KELLY)