

# **Laser Heterodyne System for Ozone Monitoring over Antarctica**

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## **Abstract**

The measurement of minor constituents along with ozone in the atmosphere is of great significance with the discovery of ozone hole phenomenon over Antarctica during spring time in particular and ozone depletion over high latitudes in general. A laser heterodyne system using a tunable CO<sub>2</sub> laser with 1 GHz Acousto-Optic Spectrometer as back end has been designed and developed at National Physical Laboratory, New Delhi to monitor vertical profiles of various minor constituents including those of ozone in the atmosphere over Antarctica. The vertical profiles can be obtained by spectral analysis of individual lines measured with ultra high spectral resolution with laser heterodyne system through inverse solution of radiative transfer equation. The inversion technique has also been developed and tested. The laser heterodyne system developed at NPL has been successfully set up at Maitri an Indian Antarctic station ( 70°46'S, 11°44'E) during 1993-94 Antarctic summer and is first of its kind over Antarctic region . The ozone line profiles obtained at Maitri are in turn used to get vertical profiles of ozone using inversion technique. In the present communication some salient features of laser heterodyne system, acousto-optic spectrometer, inversion technique and results will be discussed in detail.

## **1. Introduction**

The measurement of minor constituents in the atmosphere is of great significance to understand the physics, chemistry, dynamics and radiation budget of the atmosphere. The discovery of catalytic destruction of O<sub>3</sub> by ClO<sub>x</sub> and NO<sub>x</sub> in seventies by Johnston *et al.* [19] in general and ozone hole phenomenon [11,28] over Antarctic region during spring time in eighties in particular has generated an unprecedented surge of interest in the monitoring of ozone and other related minor constituents in the atmosphere. It is well known that our earth is shielded by a thin layer of ozone from harmful effects

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of solar ultraviolet UV-B radiation which is a primary cause of skin cancer besides its adverse impact on plants, agriculture and climate. Also now it has been recognized that the green house gas effect due to non CO<sub>2</sub> gases such as CFC's, N<sub>2</sub>O, CH<sub>4</sub>, O<sub>3</sub> etc. is as much as that due to CO<sub>2</sub> gas while it was only due to CO<sub>2</sub> gas during the preindustrial era. The increase of these green house gases in the atmosphere may cause global warming which in turn may raise the sea level globally and the impact on coastal cities and low lying countries such as Bangladesh, Netherlands, Maldives etc. would be catastrophic. Therefore the global measurement of vertical and horizontal distribution and temporal distribution of various atmospheric gases has become essential.

Infrared spectroscopy plays a great role in the measurement of atmospheric monitoring as, if not all, most of the atmospheric minor constituents have vibrational rotational characteristic absorption lines in the infrared window (8-12 μm) region. Careful study of their spectra with high resolution technique will provide a quantitative information about the concentration of various minor constituents. Infrared spectrometers have the capability to resolve individual spectral lines for example Michelson interferometers provide spectral resolving power of  $10^5 - 10^7$ . Laser Heterodyne Spectrometer have ultra high spectral resolution ( $10^5 - 10^7$ ) and quantum limited sensitivity [3,24]. Therefore laser heterodyne spectrometer can reveal complete spectral line profiles even for Doppler broadened absorption lines formed in the upper atmosphere.

Keeping this in view a tunable CO<sub>2</sub> laser heterodyne system has been designed, developed and set up at National Physical Laboratory, New Delhi to monitor various trace species in the troposphere and stratosphere. The laser heterodyne system with one GHz acousto-optic spectrometer (AOS) as back end after successful operation at NPL has been taken to Maitri, an Indian Antarctic station, 70°46'S, 11°44'E) during 1993-94 Antarctic summer and it has been operated there successfully to obtain ozone profiles and is first of its kind over Antarctic region. In the present communication some features of laser heterodyne system along with its automation using a one GHz acousto-optic spectrometer, inversion technique and results obtained are discussed in detail.

## 2. Experimental Setup

### 2.1 Laser Heterodyne System (LHS)

Infrared laser heterodyne spectroscopy provides a powerful tool for identification of weak molecular and atomic species. The advantages of the laser heterodyne system over other techniques are its ultra high spectral resolution, high spatial resolution, high quantum detection efficiency and very good signal

to noise ratio. The high resolution makes the system very selective as the interference problem due to overlapping lines or bands are minimized and the lines can be resolved completely with high resolution which outfits the other techniques.

The block diagram of the system designed, developed and set up at NPL is depicted in Fig.1. The CO<sub>2</sub> laser used as a local oscillator is tuned on a line corresponding to the absorption line of the minor constituent of the interest. The solar tracker (heliostat) follows the sun and brings in the solar radiation. The incoming chopped solar radiation is filtered out and IR radiation (8-12 μm) and CO<sub>2</sub> laser beam of moderately low power are combined via a Zinc Selenide beam splitter. The solar radiation and CO<sub>2</sub> laser beam are co-aligned and focused on the high speed liquid nitrogen cooled Hg Cd Te detector which acts as a mixer as well as narrow band filter. The detected signal is nothing but the difference frequency (IF). The IF signal is further amplified in low noise wide band (5-1200 MHz) RF amplifiers and passed through various RF filter channels (25-1200 MHz, old version not shown in Figure 1). The signal from various channels is square law detected and is fed to lock-in amplifier where synchronous detection takes place. The signal can be read directly from the lock-in amplifier or recorded on a strip chart recorder. The observations taken at different frequency channels in the wings of the absorption line of the minor constituents of interest would resolve the line completely and vertical profile can be obtained with these data using inversion technique. The number of points in the wing of the line would decide the height resolution. The system has been described in detail elsewhere [14-16]. This system was being manually operated by tuning various RF filters (25-1200 MHz) and was time consuming and subjected to errors in the measurement. In view of the above to make this system semi-automatic one, a wide band acousto-optic spectrometer has been developed as described below.

## **2.2 Acousto-Optic Spectrometer (AOS)**

Acousto optical technique for power spectral measurement has been exploited for a variety of signal analysis applications [4,12,21,25,26,29]. The spectrum analysis is one of the most useful techniques in the modern science, virtually every discipline make use of it in one form or the other. Although the architecture may vary depending upon AO signal processing function, the common ingredients in these system are an optical source may be a laser diode or visible emitting gas laser, the light modulator consisting of one or more AO Bragg devices and an array of the photodetectors. The Bragg cell also known as A.O. diffraction cell serves the key role in an acousto-optic spectrometer. It converts RF signal to ultrasonic traveling waves modulating the optical index

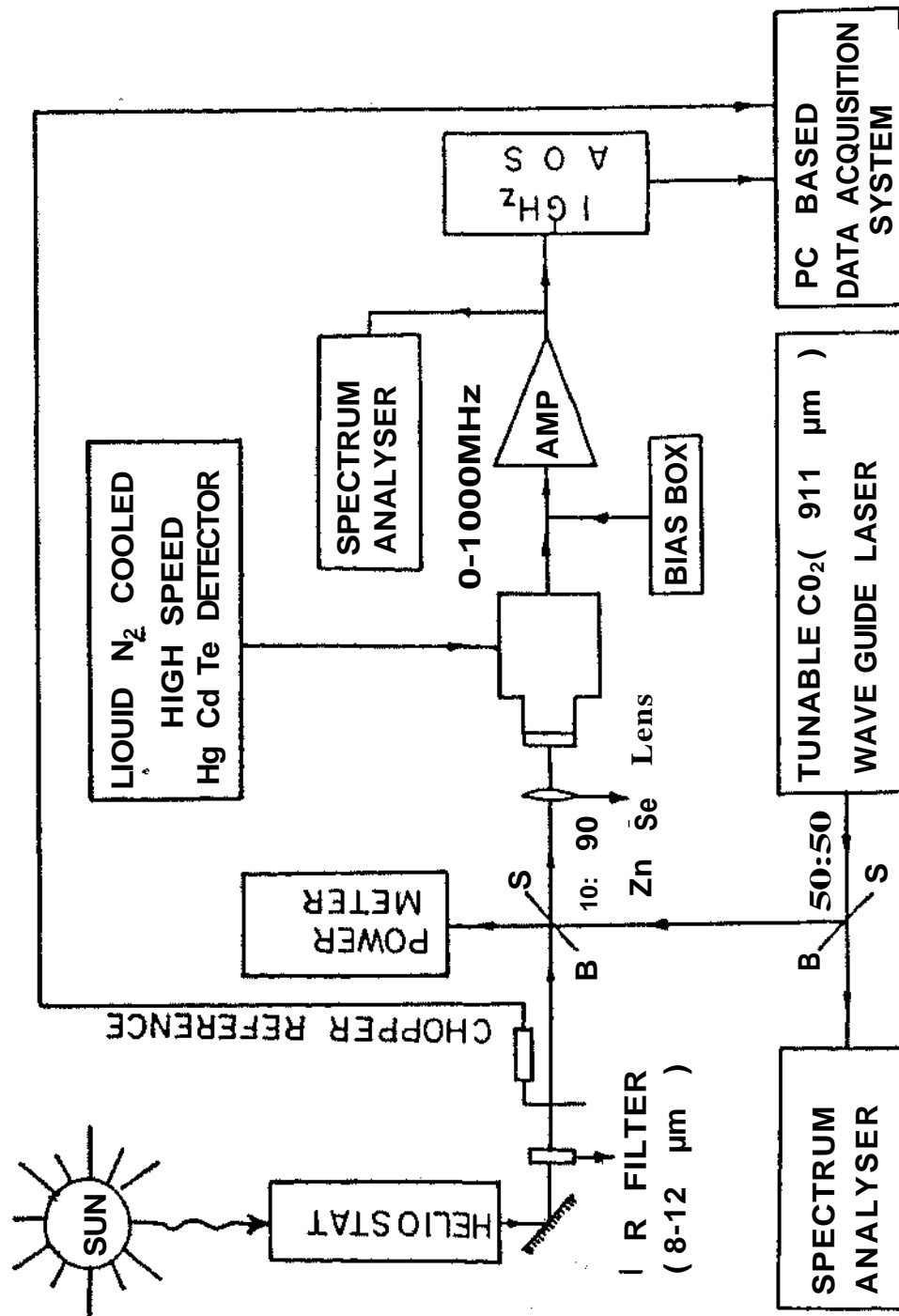


Fig.1.: Block Diagram of Laser Heterodyne System

of the cell. The cell is illuminated across its aperture by a laser beam. A fraction of light is diffracted by acoustic waves, the angle of diffraction is determined by the frequency while the intensity of the diffracted light is proportional to the power of the input RF signal. The intensity distribution can be detected by linear array of photo detectors which in turn represent the required RF power spectrum.

As described above an acousto-optic spectrometer with one GHz bandwidth has been designed and developed and was integrated with the laser heterodyne system developed at NPL, New Delhi. The block diagram of the acousto-optic spectrometer is shown in Fig. 2. The system consists of 1 GHz Bragg cell- ( $\text{Li Nb O}_3$ ), diode laser emitting at 784 nm, a Fourier lens, 1050 pixel CGD array and PC based data acquisition system. A proper software and hard ware has been developed to digitize the frequency power spectrum and stored for further analysis to get height profiles of various minor constituents.

The acousto-optic spectrometer developed has been tested with a laser heterodyne system of Reims University, France. The frequency power spectra obtained using an empty cell of 200 cm length and 600 milli torr ozone in the cell are as expected. The P(24)  $\text{CO}_2$  laser line in 9.6  $\mu\text{m}$  band was used. The spectra thus obtained shows very well defined ozone absorption lines. A spectral resolution better than 5 MHz has been obtained. A typical ozone line spectra obtained over Maitri, Antarctica during February 1994 has been depicted in Fig. 3. The use of AOS as back end of laser heterodyne system is timely and well suited to detection of weak signal buried in noise. The main advantages of the AOS are wide bandwidth, high resolution, large number of channels, high detection sensitivity and compactness, light weight and energy efficient.

### 3. Inversion Technique

The composition of temperature profiles can be obtained by analysis of individual spectral line measured at ultra high spectral resolution through inverse solution of radiative transfer equation [5-9,20,22,23] given below:

$$I_{\nu}(\mathbf{P},T) = B_{\nu}(T_s) t(\nu,\mathbf{P}) + \int_{z_1}^z B_{\nu}(T) K(\mathbf{P},T) dz \quad (1)$$

where  $I_{\nu}$  is emergent spectral intensity from a non scattering atmosphere.

$B_{\nu}(T_s)$  is the plancks function at source temperature  $T_s$

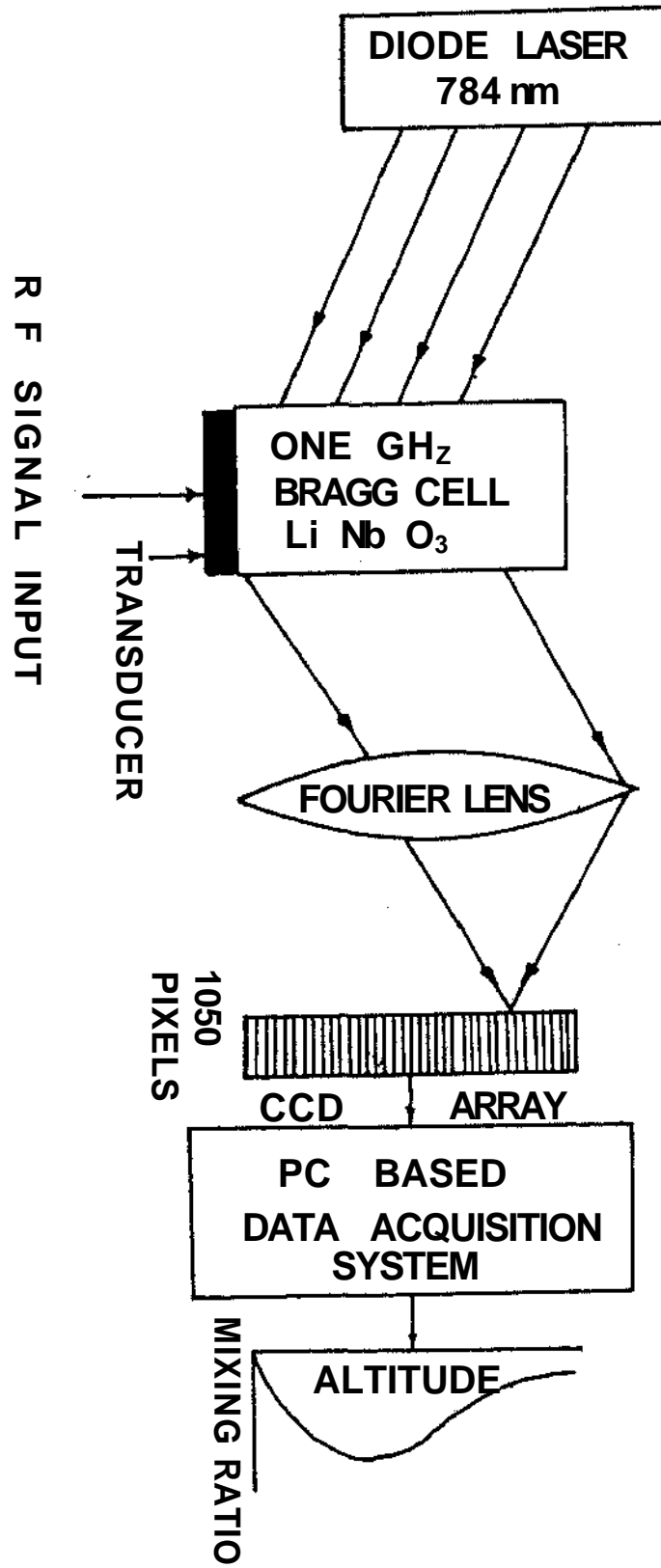


Fig.2 : Block Diagram of 1 GHz Acousto-Optic Spectrometer for Laser Heterodyne System

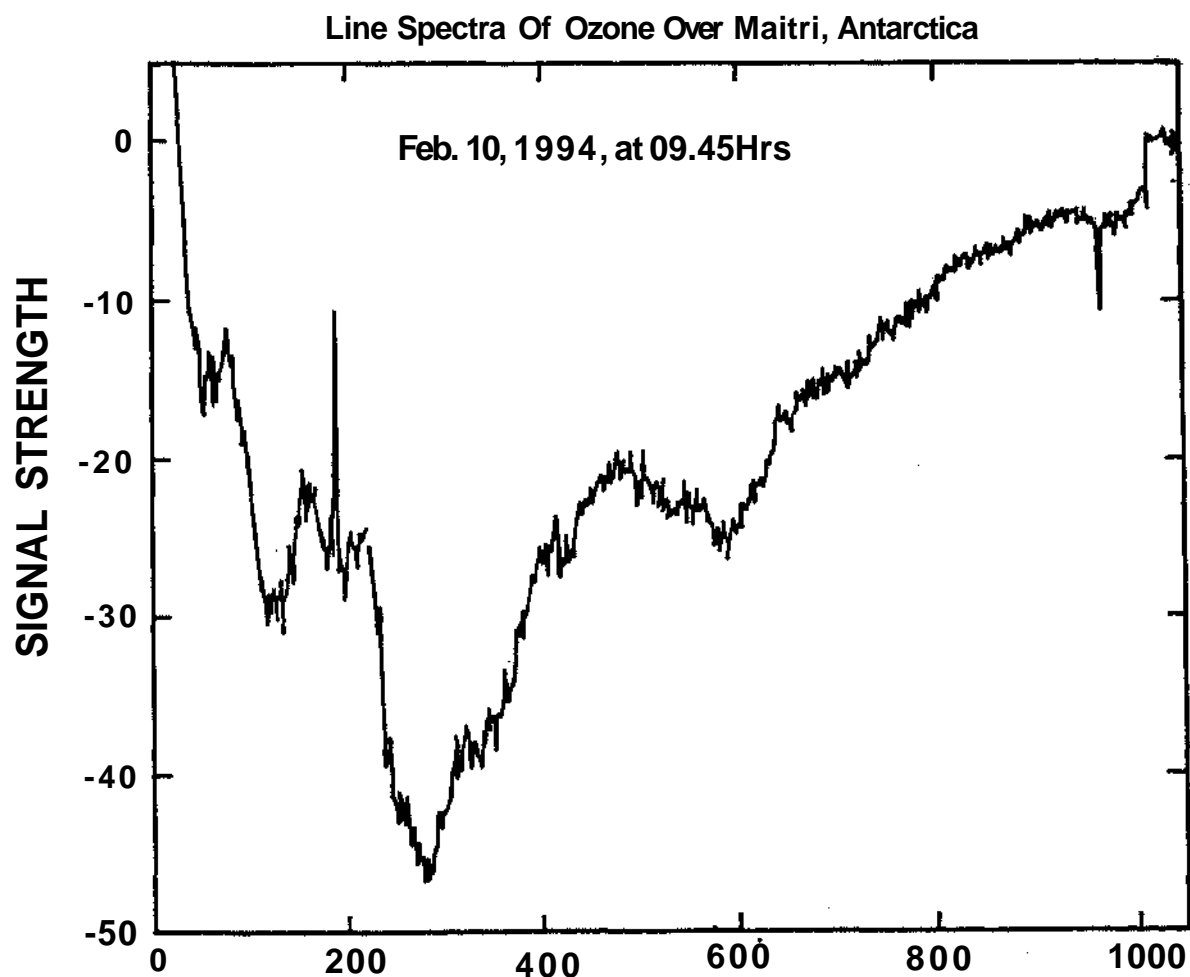


Fig.3 : Une spectra of ozone over Maitri Antarctica, obtained on Feb. 10, 1994

$(v,P)$  is the transmittance from surface to the top of the atmosphere

$K(P,T)$  is the weighing function

$$K(P,T) = d_v/dz \quad \text{and} \quad z = -\ln p \quad (2)$$

The first term in equation (1) is the surface contribution whereas the second term represents the atmospheric contribution to the spectral intensity. In the ground based measurements of the earth's atmosphere in solar absorption mode the second term i.e. self emission term in radiative transfer equations is negligible as compared to solar surface contribution and the observed spectral intensity may be written as:

$$I_v(P,T) = B(v, T_s)(v,P) \quad (3)$$

The atmospheric transmittance factor  $v$  may be given as:

$$t(\nu, P) = \exp \left[ - \int_0^L k_{\nu i} d\tau \right] \quad (4)$$

where  $k_{\nu i}$  being the specific absorption coefficient and  $d\tau$  the element of column density for the  $i$  th species of the absorbing gases given by:

$$d\tau_i(P) = q_i(P/P_0) (T/T_0) \sec X_z dz \quad (5)$$

where  $q_i$  is the volume mixing ratio of the gas,  $X_z$  is the solar zenith angle and subscript 0 refers to the reference quantities.

The spectral intensity  $I_\nu$  from planetary atmosphere is measured for an appropriately chosen set of frequencies for which weighting functions  $k_{\nu i}$  are well distributed over the atmosphere. The atmospheric composition profiles may be obtained through inverse solution of equation (2) if the temperature and pressure profiles are known.

The absorption coefficients  $k(\nu)$  at line center and in the wing of the line are calculated using the expression:

$$k(\nu) = (S/\pi) \left[ \alpha / (\nu - \nu_0)^2 + \alpha^2 \right] \quad (6)$$

where

$$\alpha = \alpha_0 = (P/P_0)(T_0/T)^{1/2} \quad (7)$$

$$S(T) = S_0(T_0/T)^{3/2} \exp\{ -1.439 E'' [(T-T_0)/T T_0] \} \quad (8)$$

where  $\alpha_0$  is the line halfwidth at  $T_0 = 298^\circ \text{ K}$

$S_0$  is the line strength at  $T_0$

and  $E''$  is lower energy level of transition

The line parameters such as  $\alpha_0$ ,  $S_0$ ,  $E''$  etc. are taken from HITRAN data base 1986 edition Rothman et al. [27]. The line shapes are generally expressed by the Voigt function which is a convolution of Lorentzian and Gaussian functions. The former is an expression for pressure broadened line shape which dominates in the troposphere and latter is that for Doppler broadened line shape which dominates in the upper stratosphere and above. In the present case below 25 km Lorentz profile was used while above 25 km Voigt profile was used. The Voigt line shapes can be expressed as:

$$k(\nu) = (S/\alpha) (y/\pi) \int_{-\infty}^{\infty} \exp [(-t^2)/(Y^2 + (x-t)^2)] dt \quad (9)$$

where



$$\alpha = (\ln 2/\pi)^{1/2} \quad (10)$$

$$x = (v - v_0) (\ln 2)^{1/2} / \alpha D \quad (11)$$

$$y = \alpha (\ln 2)^{1/2} / \alpha D \quad (12)$$

$$\alpha_D = 3.58 \times 10^7 (T/M)^{1/2} \text{ cm}^{-1} \quad (13)$$

M = molecular weight

a = collision broadening

For the present analysis a careful selection of ozone absorption line  $1043.1775 \text{ cm}^{-1}$  has been made to get sharp weighting functions and hence height resolution. The weighting functions which depend upon absorption coefficients, have a property of reaching maximum peak at different heights for different values of the frequencies i.e. at line center and in the wings of the line as shown in Fig.4 . The line parameters such as half width, line strength and lower state energy for ozone absorption line  $1043.1775 \text{ cm}^{-1}$  are given below:

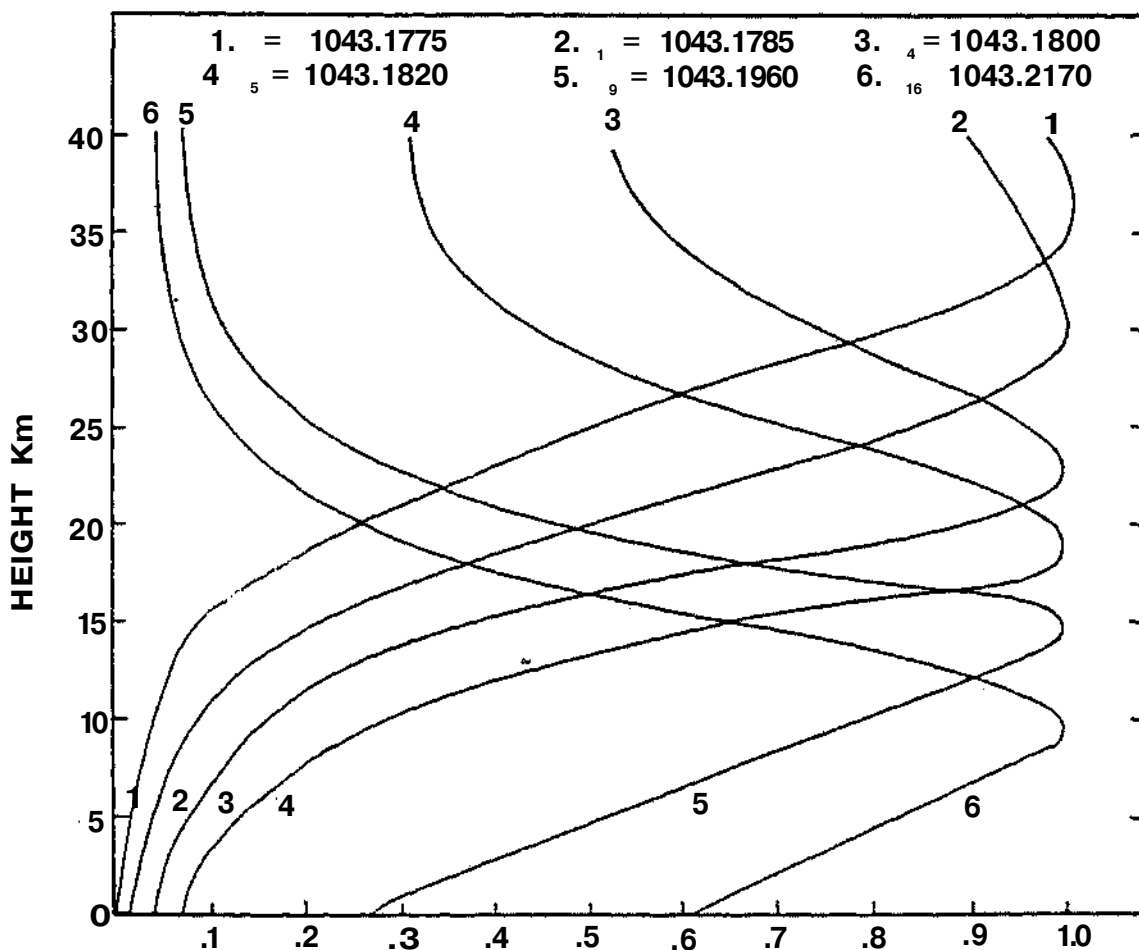


Fig.4 : Normalized weighing functions at line center  $1043.1775 \text{ cm}^{-1}$  and in the wing of this line

$$\alpha_0 = .083 \text{ cm}^{-1}; S_0 = 1.050 \times 10^{-23} \text{ cm}^{-1} \text{ atm}^{-1}; E'' = 26 \text{ cm}^{-1}$$

The line half width was calculated for this line at various heights using Lorentz, Doppler and Voigt profiles. The computations were made at line center and in the wings of the line. In all 16 channels were selected. It is to be noted that weighting functions computed peaks at different altitudes for each frequency channel and the sharpness decides the height resolution.

For retrieval of ozone profiles the actual ozone profiles obtained at McMurdo station of US A in Antarctica [ 10] are used for the present work which are representative profiles for normal and ozone hole conditions. The spectral intensity ( $D_v$ ) for each channel was computed using the above said McMurdo ozone profiles and line parameters for  $1043.1775 \text{ cm}^{-1}$ . The computed spectral intensity ( $D_v$ ) at various frequency channels is the contribution of ozone at different altitudes and is used to retrieve the height profiles for ozone using the iterative inversion method first suggested by Chahine [6] for remote sensing of atmospheric temperature by satellite observations and later developed by Abbas *et al.* [ 1,2], Jain [ 13, 17] for remote sounding of atmospheric minor constituents using high spectral resolution techniques. The inversion is carried out in following steps:

1. To start with a uniform vertical distribution of mixing ratio of ozone say 1 ppm is guessed and the spectral intensity  $I_v$  for each channel is computed corresponding to initial guess profiles.

2.  $I_v$  and  $I_v$  are compared and residual  $R^n$  is computed by

$$R^n = (I_v - I_v)/I_v \quad (14)$$

where  $I_v$  is the spectral intensity computed from the actual ozone profiles for various frequency channels for McMurdo station [10] as explained earlier. If the residual is zero or of the order of noise level of the measurement the initial guess is the solution.

3. If the residual is not zero, an improved mixing ratio at levels corresponding to the peaks of absorption coefficients is obtained from the relaxation equation:

$$q^{n+1}(P_j) = \alpha_{jq^n}(P_j) \quad (15)$$

where  $q^n$  and  $q^{n+1}$  are the  $n^{\text{th}}$  and  $n+1^{\text{th}}$  guesses,  $J$  is the scaling factor and  $P(j)$  is the pressure at level  $j$ .

4. The iteration process is repeated until each scaling constant approaches unity which is equivalent to satisfying the residual given by Equation (15).

### 3.1 CASE 1: Normal Ozone Profile

In this case the ozone profiles over McMurdo station Antarctica (78° S) obtained on Aug. 23,1989 by Deshler *et al* [10] was used for testing the inversion technique developed at NPL. The retrieved profile obtained after 20 iterations is shown in Fig.5. The retrieved profile compares well with that of original assumed profile. The inversion technique was further tested with different initial guess profile and each time the retrieved profile compared well with the original one which demonstrated that the retrieved profiles is independent of the initial guess profile. The complete profile is obtained with appropriate interpolation and extrapolation.

### 3.2 CASE 2 : Ozone Hole Condition

The sensitivity of the inversion technique during very low values of ozone in 12-22 km height range due to ozone hole developed is also tested. For these computations the ozone profile obtained again at McMurdo station on October 20,1989 (Deshler *et all* [10]) is used. The similar method for retrieval of ozone profile as explained above was used for ozone hole conditions and it is found that after 20 iterations the retrieved ozone profile compares well with that of original as shown in Fig.6. In this case also the technique was tested for different initial guess profiles and found to be independent of it.

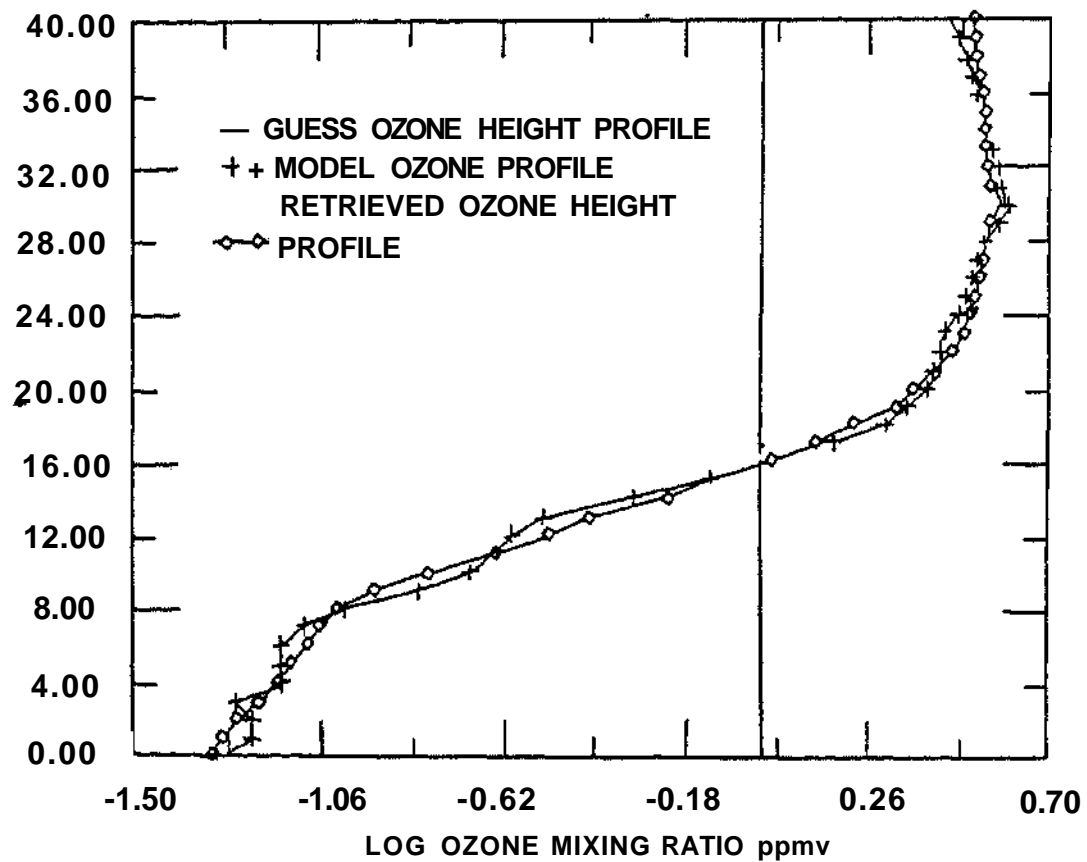
The above analysis shows that laser heterodyne system provides an important tool for monitoring the ozone height profiles using inversion technique during both normal and ozone hole conditions over Antarctica.

## 4. Results and Discussions

Two of us (Dr. S.L. Jain and Dr. Patrick Dierich) along with the 13<sup>th</sup> Indian Scientific Antarctic Expedition departed from Goa by Stepan krashennikov a Russian vessel for Antarctica on December 8,1993 and reached to Antarctica on Dec. 30, 1993. The Maitri station was first visited by one of us (Dr. S.L. Jain) along with the leader Shri Sudhakar Rao to finalise the location of the laser heterodyne hut by very first sortie on 31st Dec. 1993 and returned to the ship after the visit and site selection.

### 4.1 Hut Construction

It was planned to helilift the stores of hut by sortie no. 9 to 29 on priority basis but may be due to some operational problems it was rescheduled to be helilifted after all the other scientists being inducted. Even this could not be done by 9-1-94 and seeing the situation we requested the leader to take the help

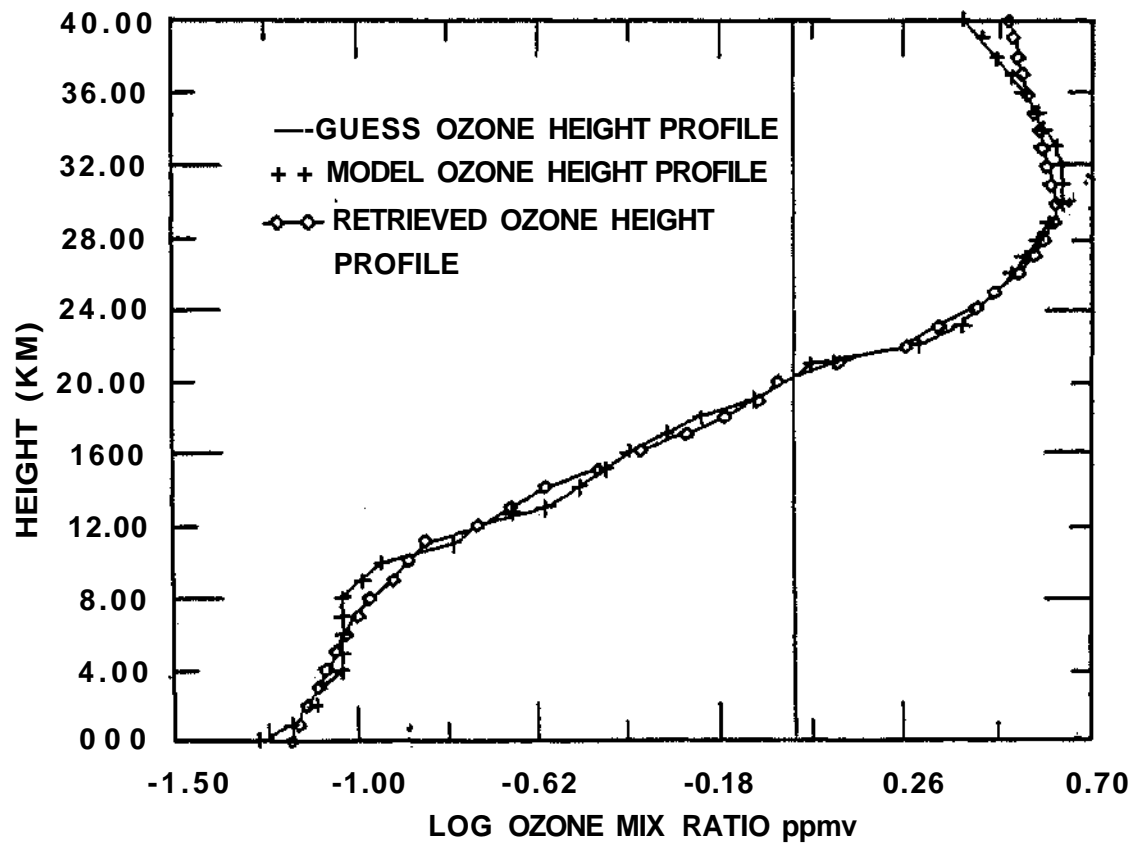


*Fig.5: Retrieved Antarctic ozone height profile normal case*

of Russian MI-8 helicopter or the Russian convoy to send the stores to Maitri and leader agreed to load the hut stores in Russian sledge which was done on 11-1 -94. One of us (Dr. S.L. Jain) was also inducted to Maitri on 11-1-94. The hut stores reached Sanklap point on 13-1-94 from where it was helilifted on 15-1 -94 and 16-1 -94. With the help of army team the hut was erected and was complete in all respects including electrical wiring and electronically controlled heating facility on 24-1-94. The construction work was done in the harsh Antarctic environmental conditions on war level

## 4.2 Laboratory Setup

The laboratory set up started on 25-1 -94 with testing of all the individual instruments. With in next four days the system was aligned and tested with black body source and laser heterodyne signal was obtained using black body source at 1373° K for the first time over Antarctic region and system was ready to test with solar radiation using heliostat which was already installed and its tracking was tested during hut erection. Due to want of cloudless clear sky we could not start operation of the system with solar radiation till 6-2-94 There was a great excitement when for the first time real laser heterodyne signal using

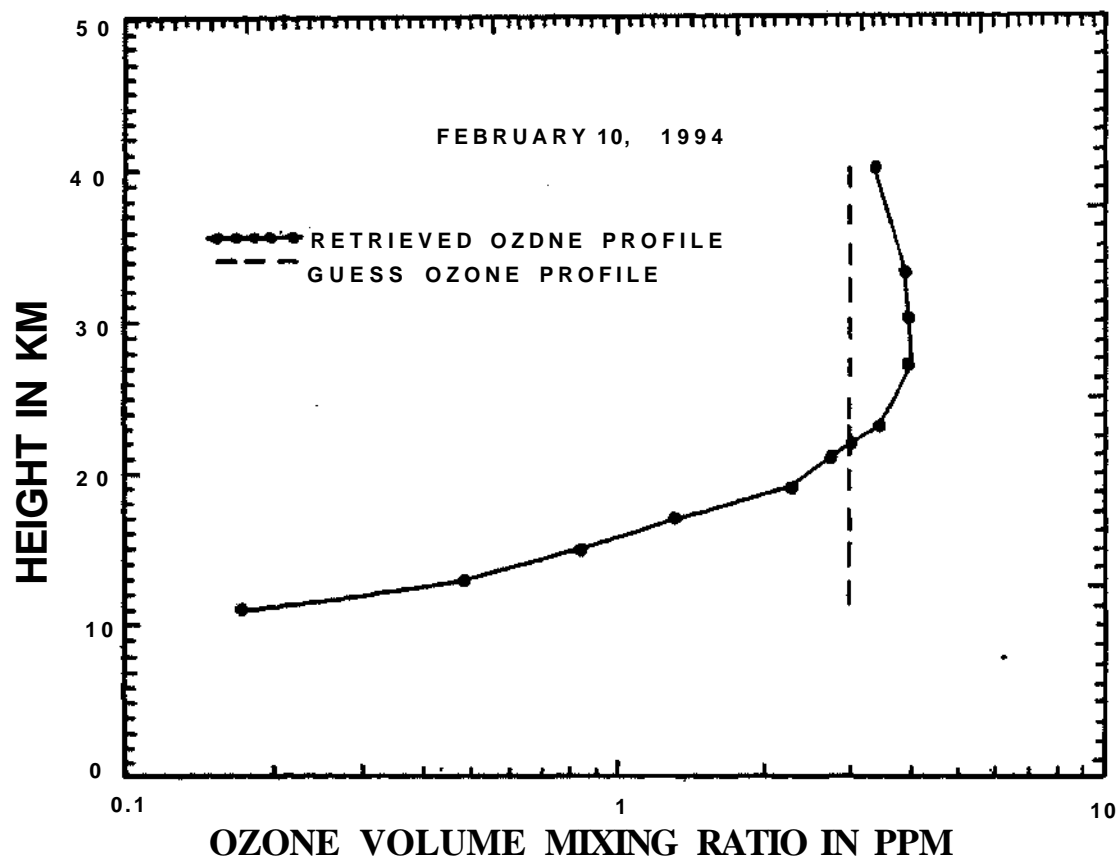


*Fig.6: Retrieved Antarctic Ozone Height Profile Ozone Hole Case*

solar radiation and CO<sub>2</sub> laser emission at P(24) line was obtained on 6- 2-94 and ozone line profile was observed again for the first time over this region. The data collection was continued on all clear blue sky days.

The absorption line selected is 1043.1775 cm<sup>-1</sup>. A typical ozone line spectra obtained over Maitri is shown in Fig. 3. The line spectra thus obtained has in turn been used to get ozone vertical profile using inversion technique. A guess profile of uniform mixing ratio of 3ppm as shown in Fig. 7 has been used for the retrieval. A typical retrieved ozone profile from the line spectra obtained on February 10,1994 is shown in Fig. 7.

The laser heterodyne system has thus demonstrated successfully its capability to monitor ozone height profiles in the atmosphere. The efforts are on to extend the facility to monitor other constituents to understand the complex interaction between atmospheric dynamics, chemistry and radiation budget which in turn requires a large data base on regular basis at tropical as well as at Antarctic latitudes. In order to extend the wavelength coverage of the CO<sub>2</sub> laser the other carbon isotopes such C-14 or C-13 are to be used which in turn needs some R and D work. The above analysis shows that laser heterodyne system provides an important tool for monitoring the ozone height profiles



*Fig.7: Ozone volume mixing ratio at Maitri, Antarctica*

using inversion technique during both normal and ozone hole conditions over Antarctica. The accuracy of the inversion technique will depend to a great extent on the accuracy of the line parameters which still needs the extensive research work. The present system can not be used during night and cloudy days as it uses sun as the source. However, it can be modified to use it during such conditions by using it in emission mode instead of absorption mode.

### 43 Liquid Nitrogen

Enough liquid nitrogen in IBP made cryocans TA-55 was brought from Goa to carry out the experiment during the summer at Maitri and even after evaporation on the way 50 % of the liquid nitrogen was available for the use. The cryocans are of special type to hold the liquid nitrogen more than two months.

We also brought liquid nitrogen plant to install and put it in operation for the future at Maitri. The capacity of the plant is 1 liter of liquid nitrogen per hour. But due to want of proper room, it could not be installed during the expedition and was stored duly packed in a metallic container for its installation in due course of time.

## 5. Problems Experienced

1. For the safety of the sensitive and expensive instruments like ours the perfect electrical earthing is must and expert opinion may be sought for the same keeping in view the special Antarctic conditions ( per ma frost, low humidity etc). At the first instance we observed 50-60 volts between earth and neutral which was brought down to 20-30 volts by our efforts.

2. HF interference is very strong and maybe we have to do some shielding for the same.

3. A major component ie secondary mirror of the heliostat got damaged due to low temperature outside the laboratory and needs replacement with better protecting coating.

4. During the HF communication high voltage fluctuations are observed and it is suggested that separate generators should be used for experimental work and HF transmission and it may be needed to stop HF broadcast during observing time.

5. With the first hand experience with Antarctic conditions we strongly felt the need of some modifications in the system and the software as well as hardware. Therefore the major and sensitive components of the system were taken back for necessary modification.

6. Electrostatic charge is an another problem and one have to get discharge himself before touching any sensitive instrument.

7. The availability of very few cloudless clear sunny days limits the regular observations by this system.

## 6. Acknowledgement

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