

A-1.1.3 Studies on Development of Retrieval Method of Atmospheric Minor Constituents by the Infrared Heterodyne Spectroscopy

Contact person Hiroshi Fukunishi
Professor
Department of Astrophysics and Geophysics
Faculty of Science, Tohoku University
Aramaki Aoba, Sendai 980-77 Japan
Tel: +81-22-217-6734 Fax: +81-22-217-6739
E-mail: fuku@stpp2.geophys.tohoku.ac.jp

Total Budget for FY1994-FY1996 13,711,000 Yen (FY1996; 4,364,000 Yen)

Abstract

Observations of infrared absorption lines of nitrous oxide were made with a laser heterodyne spectrometer in Sendai in 1994. A retrieval method to obtain vertical profiles and total column densities of nitrous oxide was developed and errors in the retrieval were precisely examined by computer simulations. From observations by a new laser heterodyne spectrometer in Antarctica, we have obtained high-resolution infrared spectra of ozone, nitrous oxide, nitric acid and methane. A rapid variation of ozone mixing ratio in the stratosphere during a growth phase of the Antarctic ozone hole is found from consecutive observations with a high time resolution of 10 minutes.

Key Words Atmospheric Minor Constituents, Infrared Spectroscopy,
Laser Heterodyne, Spectrometer, Remote Sensing, Ozone Hole

1. Introduction

Atmospheric minor constituents such as water vapor, carbon dioxide, ozone, etc. exhibit absorption bands in the infrared region which is one of the atmospheric windows. Each absorption band consists of many absorption lines due to rotation-vibration transitions. A line profile contains information on atmospheric temperature, pressure and concentration of absorbing molecules themselves. Therefore, vertical profiles of density distribution of those minor species can be derived from high-resolution spectra of infrared absorption lines.

Our group in Tohoku University has been developing laser heterodyne spectrometers for remote sensing of atmospheric minor constituents from the ground [Taguchi et al., 1994a]. The laser heterodyne spectroscopy enables us to measure infrared spectra with an ultra-high spectral resolution and quantum-limited sensitivity. We have succeeded in observations of vertical profiles and total column densities of ozone, nitrous oxide and methane with the laser heterodyne spectrometers [Koide et al., 1995a].

In this study, we develop a new laser heterodyne spectrometer for observation of the Antarctic ozone hole and related minor species. An inversion method to retrieve vertical profiles of nitrous oxide is developed and error sources are precisely examined by computer simulations.

2. Instrumentation

A portable laser heterodyne spectrometer (TDLHS-3) was developed for observation of the Antarctic ozone hole [Taguchi et al., 1994b]. A schematic diagram of TDLHS-3 is shown in Figure 1. This spectrometer has following characteristics:

- 1) Four diode lasers are installed in it as a local oscillator. They operate in different wavenumber regions from 900 through 1220 cm^{-1} .
- 2) Optical alignment can be easily achieved and stability is high.
- 3) A gas absorption cell is equipped to identify weak absorption by, for example, nitric acid.

The local oscillator lasers are cooled by liquid nitrogen to 65-90K temperatures. Stability of the laser temperature is important for stable oscillation. The laser temperature can be stabilized within 0.01K. Each local oscillator laser emits in a 50 cm^{-1} wavenumber region. By switching four lasers, four atmospheric minor species, nitric acid, ozone, nitrous oxide, and methane, are observed in the wavenumber regions of 900, 1100, 1190, and 1220 cm^{-1} , respectively.

The sun is used as a light source of the laser heterodyne observations. The solar beam is automatically tracked by a sun follower, and chopped by an optical chopper for phase-sensitive detection. A required narrow wavenumber region in the solar infrared radiation is selected by a diffraction grating. The laser beam and the solar beam are combined at a beam splitter, and fed into a high-speed infrared detector. Heterodyne signal from the detector is amplified, filtered and square-law detected, and the output is acquired by a computer, which is also used for control of the laser current.

3. Observations and Results

3-1. Observations in Japan

We initially planned to observe nitric acid with the laser heterodyne spectrometer, however, we gave up this plan because of failure of local oscillator lasers operating in the 900 cm^{-1} region where an absorption band of nitric acid exists. Observations of nitrous oxide were made with TDLHS-2 in Aobayama Campus of Tohoku University in Sendai. An absorption line of nitrous oxide at 1182.6469 cm^{-1} is observed. An example of observed absorption lines of ozone and nitrous oxide is shown in Figure 2. The observation of nitrous oxide started in May 1994 and made on 19 days until December 1994.

Vertical profiles of mixing ratios of nitrous oxide in the altitude range of 5-30km are derived from the observed absorption spectra by an inversion method [Shigematsu et al., 1995]. Figure 3 shows a vertical profile of nitrous oxide obtained from the observation with TDLHS-2. A monthly average temperature in Sendai is used for the inversion calculation. Absorption line parameters are taken from the HITRAN database 1992 edition [Rothman et al., 1992]. The retrieved values are consistent with nitrous oxide concentration measured by remote sensing from a satellite and sampling by balloons.

Errors in the inversion calculation are evaluated by computer simulations. For systematic errors, 3.0% error at 25km altitude for the initial vertical profile of nitrous oxide, 1% error at 30km for the temperature model, 1.9% error at 30km altitude for the pressure model, and 0.9%, 9.2% and 2.1% errors for the line intensity, pressure-broadened half-width and line position, respectively, are estimated. For random errors, 16.9% error at 30km altitude is estimated when a signal-to-noise ratio (S/N) of an observed spectrum is 100. This error decreases to 3.4% when S/N is 500.

3-2. Observations in Antarctica

TDLHS-3 was brought into Syowa Station in Antarctica in January 1994. Several test observations were made before beginning of the polar night. Observations of ozone and nitric acid were started in late August. Nitrous oxide was sometimes observed when fine weather continued several days. Observations of methane were made in November and December 1994. Figure 4 shows representative spectra of ozone, nitrous oxide, methane and nitric acid obtained in Syowa Station in 1994.

We have analyzed ozone data so far, and obtained temporal variations in vertical profiles of ozone with a time resolution of 10 minutes for the observations in September and October [Koide et al., 1995b]. The Antarctic ozone hole is clearly seen in these data. There can be seen a large variation above 20km altitude in some days. Figure 5 shows such a temporal variation in vertical profiles of ozone observed on September 26, 1994. To guess the cause of this variation, we calculated potential vorticity from the objective analysis data by the Japan Meteorological Agency. It is found that Syowa Station was on the edge of the polar vortex when a large variation of ozone vertical profiles was observed. Thus, these variations may be related to an air exchange process across the boundary of the polar vortex.

For the other species, we are now proceeding analysis of the obtained data. Unfortunately, absorption due to nitric acid is too small to retrieve vertical profiles. We have to develop a least-square fitting method to derive column amounts of nitric acid from the observed absorption spectra.

4. Conclusions

Observations of nitrous oxide were made in Sendai, Japan. We developed an inversion method to retrieve vertical profiles of nitrous oxide from the observed spectra, and evaluated errors in the inversion calculation. From the observations with TDLHS-3 in Antarctica, intense variations of ozone in the altitude above 20km were found. These variations may reflect an air exchange process across the boundary of the polar vortex. We hope these progresses in TDLHS observations and data analysis lead to future application of the laser heterodyne spectroscopy from balloon or satellite altitudes for global monitoring of the atmospheric environment.

References

- Koide, M., M. Taguchi, H. Fukunishi, and S. Okano, Ground-based remote sensing of methane height profiles with a tunable diode laser heterodyne spectrometer, *Geophys. Res. Lett.*, 22, 401-404, 1995a.
- Koide, M., M. Taguchi, H. Fukunishi, and S. Okano, Observations of ozone, methane, nitrous oxide, and nitric acid with a laser heterodyne spectrometer at Syowa Station in Antarctica, Abstract for SGEPS fall meeting in 1995, B22-08, 1995b.
- Rothman et al., The HITRAN molecular database: Editions of 1991 and 1992, *J. Quant. Spectrosc. Radiat. Transfer*, 48, 469-507, 1992.
- Shigematsu, N., M. Taguchi, S. Okano, and H. Fukunishi, Observations of vertical profiles of stratospheric nitrous oxide with a laser heterodyne spectrometer, Abstract for SGEPS spring meeting in 1995, pp.559, 1995.

Taguchi, M., S. Okano, M. Koide, N. Shigematsu, and H. Fukunishi, Remote sensing of atmospheric minor constituents by the infrared laser heterodyne spectroscopy, Proceedings of 8th International Symposium on Solar Terrestrial Physics Part I, pp.206, 1994a.

Taguchi, M., S. Okano, M. Koide, N. Shigematsu, and H. Fukunishi, Development of tunable diode laser heterodyne spectrometers for remote sensing of atmospheric minor constituents, Abstract for 17th International Laser Radar Conference, 259-260, 1994b.

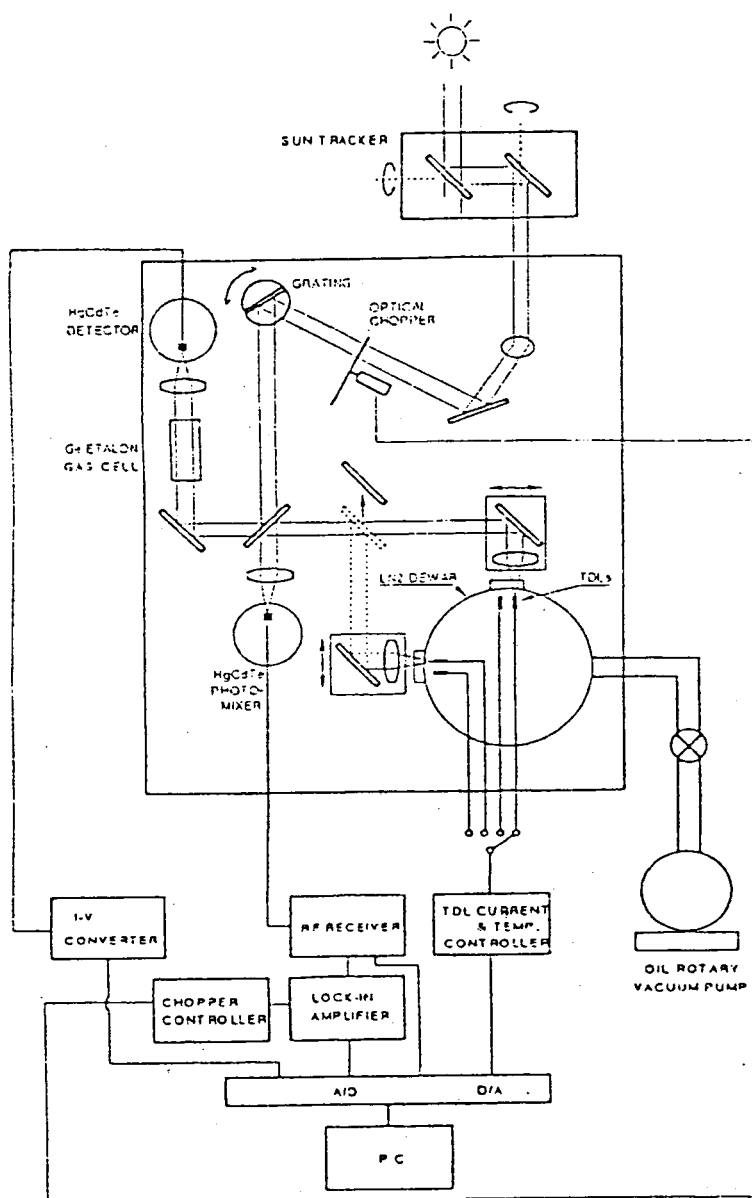


Fig. 1 A schematic diagram of the developed laser heterodyne spectrometer (TDLHS-3).

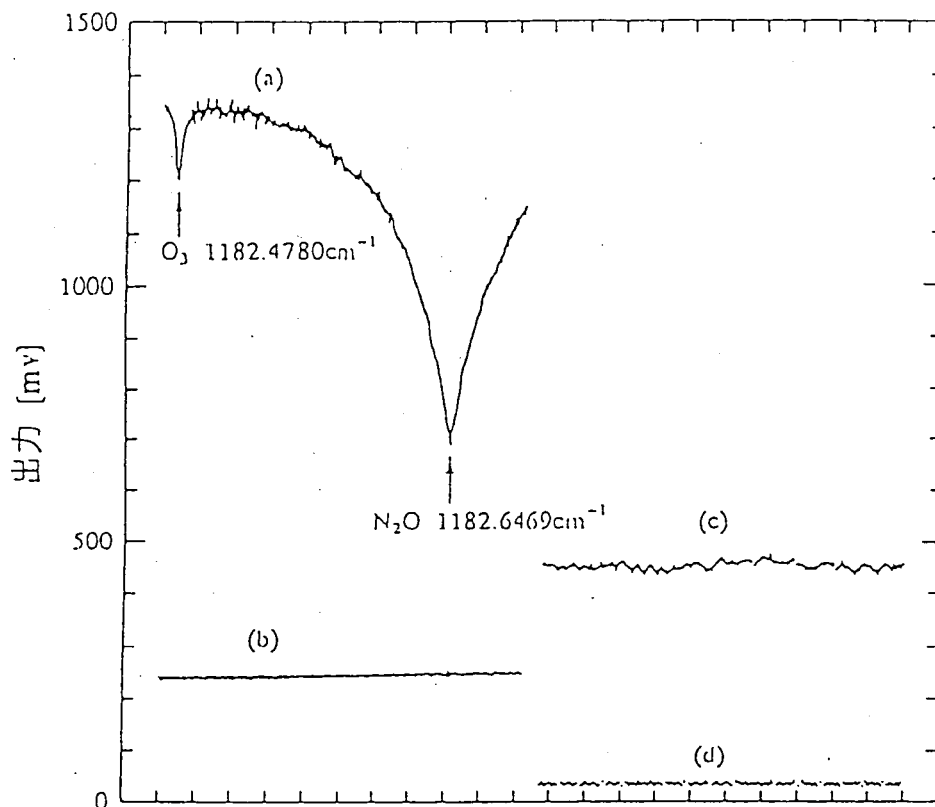


Fig. 2 An example of infrared absorption lines of ozone and nitrous oxide observed with the laser heterodyne spectrometer in Sendai on May 18, 1994. Trace (a) is the heterodyne signal, trace (b) shot noise due to the signal, trace (c) DC photocurrent due to the local oscillator and signal, and trace (d) DC photocurrent due to the signal.

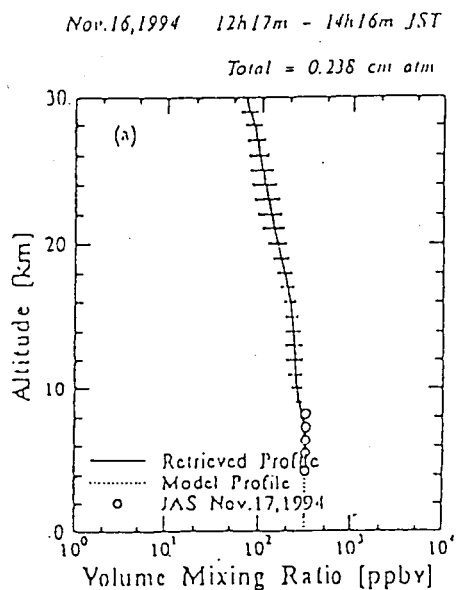


Fig. 3 A vertical profile of volume mixing ratio of nitrous oxide observed in Sendai on November 16, 1994. The results from an airborne sampling experiment are also shown by circles for comparison [Nakazawa, private communication].

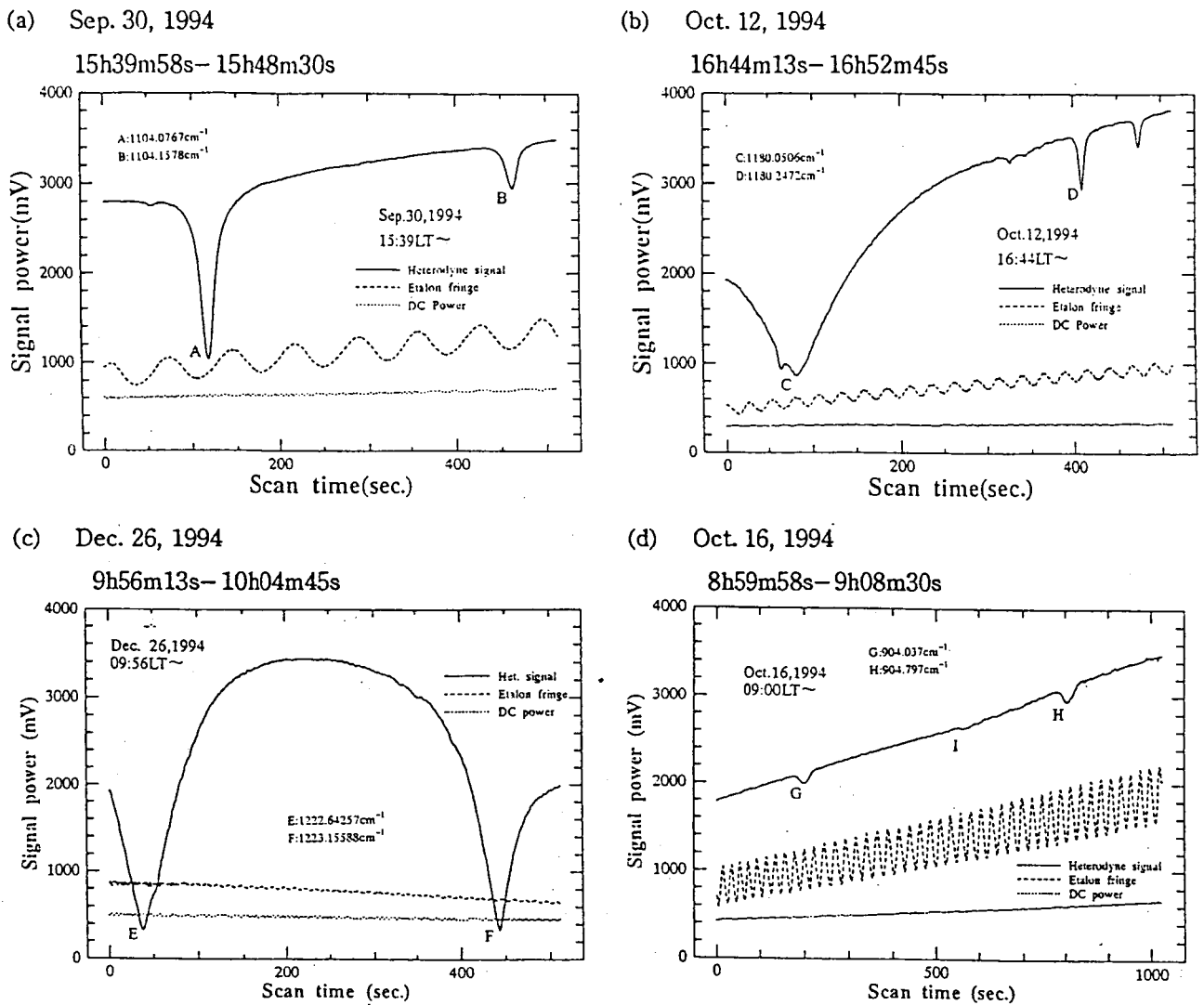


Fig. 4 Examples of absorption lines of (a) ozone, (b) nitrous oxide, (c) methane, and (d) nitric acid observed in Syowa Station, Antarctica. The times are in local time (LT=UT+3h).

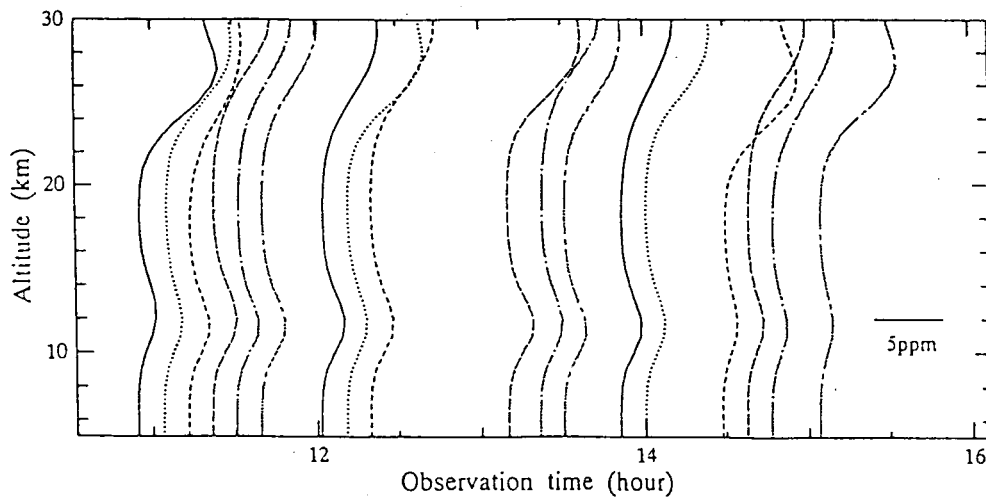


Fig. 5 Temporal variations of vertical profiles of ozone mixing ratio observed in Syowa Station on September 26, 1994. Ozone is depleted in the altitude range of 14–22km by the Antarctic ozone hole. An intense variation is seen in the altitude region above 20km.