4-6 Trace Gas Observation with Poker Flat FTIR

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We have been observing atmospheric trace gases using Fourier transform infrared spectrometer (FTIR), which was installed at Poker Flat observation site, Alaska (65.07˚N, 147.26˚W) since July 1999. FTIR has often been used to derive the total column amount of trace gas from its intensity of the absorption spectrum. In this paper, we have derived height profile of ozone concentration from line shape of the absorption spectrum of ozone using a retrieval method. We explain the scheme of the retrieval method we used and the comparison between our results retrieved and those obtained from other instruments.

Keywords

FTIR, Trace gas, Height profile, Retrieval method, SFIT2

1 Introduction

In addition to its main components—nitrogen, oxygen, and water vapor—the Earth's atmosphere contains atmospheric trace gases, such as ozone and carbon dioxide, at extremely low abundance. The volume mixing ratios of these gases to the atmosphere are extremely low, at 10^{-2} to 10^{-9}, but the gases play key roles in phenomena such as ozone depletion and global warming. By studying the trends in the abundance of the gases, we may be able to gain insights into the various phenomena occurring in the Earth's atmosphere.

Since an atmospheric trace gas absorbs electromagnetic waves at characteristic wavelengths, sunlight and moonlight will display different transmittance for different wavelengths when passing through the atmosphere. Therefore, the abundance of a given atmospheric trace gas can be determined by measuring the absorption of electromagnetic waves at the characteristic wavelength of the trace gas. Furthermore, since a trace gas in an excited state emits electromagnetic waves at characteristic wavelengths, it is also possible to determine its abundance by observing the thermal emission spectrum.

As part of the Alaska Project, an international collaboration between the Communications Research Laboratory (CRL) and the University of Alaska Fairbanks (UAF), observations of atmospheric trace gases in the polar atmosphere have been conducted using the Fourier transform infrared spectrometer (FTIR) at the UAF's Poker Flat Research Range (65.07˚N, 147.26˚E). Abundance of atmospheric trace gases from the troposphere to the stratosphere (in an altitude range from approximately 0 to 30 km) can be determined by FTIR observations.

Observations of infrared solar absorption spectra and thermal emission spectra of atmospheric trace gases have been performed with FTIR installed at Poker Flat (referred to as the Poker Flat FTIR) since July 1999. FTIR splits the light entering the spectrometer into two paths. One path is reflected by a fixed mirror and the other is reflected by a moving mirror. The interference of these two light beams is measured with the detector. The change in the intensity of the interference of light with respect to the position of the moving mirror (i.e., the interferogram) is converted numerically by Fourier transform to produce an infrared spectrum[1]. A ground-based FTIR can perform simultaneous observations of many kinds of trace gases, since it is capable of measuring a wide range of wavelengths at once. Further, it is relatively easy to examine long term variations in the atmosphere by FTIR observations, since FTIR is in a more advanced state of development than instruments such as the millimeter-wave radiometer. In particular, gases concerning global warming such as carbon dioxide, methane, ozone, and water vapor absorb infrared light, so FTIR is especially suited for observation of these gases.

In this paper, we will describe the methods of determining the abundance of atmospheric trace gases based on the infrared solar absorption spectrum observed with FTIR. Conventional FTIR observations focused mainly on determining the total column amount of a trace gas from the intensity of its absorption spectrum[2][3]. In recent years, however, it has become increasingly important to obtain information on the abundance of trace gases in each altitude range, to gain a deeper understanding of the chemical and transport processes in the troposphere and the stratosphere. Development thus began in 1995 of an algorithm to determine the vertical profile of the abundance (height profile) of a trace gas from the line shape of the absorption spectrum using the retrieval method. In particular, the SFIT2 program—developed jointly by the National Institute of Water and Atmosphere (NIWA) of New Zealand and the U.S. National Aeronautics and Space Administration's Langley Research Center (NASA Langley)—has been the most widely used for data analysis[4][5]. In the past, few studies have been conducted in Japan using the retrieval method. Presently, we are promoting the introduction of the SFIT2 program with the cooperation of the National Institute for Environmental Studies and Tohoku University, and efforts are currently underway to adjust the conditions of analysis and to determine height profiles.

This report discusses ozone height profiles obtained from ozone absorption spectra observed for the nine-month period from February to October 2001 at Poker Flat, followed by comparisons and validations made with reference to results gained through other means of observation. Ozone is a relatively abundant trace gas in the atmosphere, and in the past it has been observed and verified using instruments other than the FTIR[6]. Thus it is suited to be used for comparison and validation studies of FTIR results.

2 Observation Methods

The Poker Flat FTIR has performed virtually continuous remote-controlled automatic observations since it was installed in July 1999. The resultant collection of observation data will certainly inform discussions of variations in trace gases over various time scales longer than several days.

The high-speed network system between Alaska and CRL (referred to as SALMON, or the System for Alaska Middle-atmosphere Observation data Network) has greatly contributed to automatic observation using the Poker Flat FTIR. The daily observation schedule can be preset via the Internet, and the observation will be conducted automatically according to the schedule. SALMON enables us to automatically transfer the observed spectrum to a server inside CRL once every 30 minutes.

In ground-based observations, large changes in the solar zenith angle during measurement will result in a significant shift in the observed atmospheric region. Therefore, a single measurement is desired to be kept within 10 minutes. However, information on the line shape of the absorption spectrum is essential when determining the height profile of a trace gas with the retrieval method, and so both the frequency resolution and the signal-to-noise ratio must be sufficiently high for spectral observation.

To obtain a spectrum with a high frequency resolution, it is necessary to increase the amount of shift of the moving mirror; this requires some time. The spectrometer used in the Poker Flat FTIR (Bruker 120HR) has a maximum frequency resolution of 0.0019 cm^{-1}, but to achieve this resolution, a single cycle of the moving mirror will require 10 to 20 minutes. Moreover, the number of cycles made by the moving mirror must be increased to obtain spectra with high signal-to-noise ratio, and the observation time will increase according to the number of cycles. In the case of the ozone-absorption spectrum (near 3051 cm^{-1}) used in the present study, the Doppler broadening of the spectrum due to thermal motion of ozone in the atmosphere is larger than 0.004 cm^{-1}, and thus it was concluded that there would be no significant differences between measurements taken at frequency resolutions of 0.0035 cm^{-1} and 0.0019 cm^{-1} when obtaining the height profile. Therefore, the frequency resolution of the spectrum observed by the Poker Flat FTIR was reduced to 0.0035 cm^{-1} to shorten the time required for a single cycle of the moving mirror, and the number of cycles was doubled to improve the signal-to-noise ratio of the spectrum.

Since observation began with the Poker Flat FTIR in July 1999, daily observations were conducted from April to October 2000 and from February to October 2001 under the conditions described above. Five to ten spectra were obtained per day on days satisfying conditions such as clear sky. In this analysis, spectra with signal-to-noise ratios high enough to allow significant extraction of information were obtained for 73 days out of the 247 days during the period from February 24 to October 28, 2001.

3 Analysis Method

Previous studies have focused mainly on using the intensity of the absorption spectrum of the target trace gas to obtain the total column amount of the gas using the conventional forward method for determining abundance of the gas based on infrared absorption spectra observed using FTIR. Most of these studies have used the SFIT program developed by NASA Langley[2][3]. In SFIT, an assumption is made regarding the vertical profile (height profile) of the target trace gas, and the assumed profile is multiplied by a factor to calculate a theoretical spectrum with the smallest residual compared to the observed spectrum using the least-squares fitting method to determine the total amount of the target gas in a vertical section (total column amount). Since changing the height profile of a trace gas will increase the number of parameters optimized in the least-squares fitting method and will thus produce an infinite number of solutions with a residual of zero, height profiles cannot be determined by SFIT. Moreover, the obtained total column amount will only be valid when the assumed height profile is consistent with actual atmospheric conditions. When this is not the case, the margin of error tends to be larger than with the retrieval method described below.

In contrast, the retrieval method produces a height profile for a trace gas based not only on the intensity but also on the line shape of the absorption spectrum[4][5]. Furthermore, a better estimation of the total column amount can be made than with the forward method. As shown in Fig.1, the infrared absorption spectrum for trace gas observed with ground-based FTIR is a superposition of the absorption spectra at different altitude levels between the sun and the earth. Here, pressure and Doppler broadenings will be present in the line shape of an absorption spectrum at a certain altitude, depending on the temperature and pressure. The Doppler broadening does not significantly change with altitude, but pressure broadening decreases rapidly with increasing altitude, since it changes with atmospheric pressure. As a result, in the altitude range at which the effect of pressure broadening is broader than Doppler broadening, the spectrum will have a wide line shape at low altitudes and a narrow line shape at higher altitudes. The retrieval method uses such differences in the line shape at each alti-

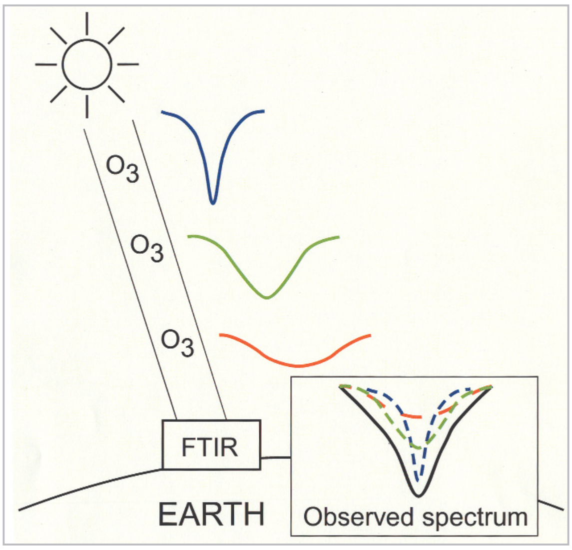

Fig.1 *A schematic diagram of the infrared absorption spectrum observed by FTIR*

The observed spectrum is a superposition of the absorption spectra at different altitude levels from the sun to the ground. The retrieval method uses the difference in the line shape at each altitude level to determine the abundance of a trace gas at each level (height profile).

tude to determine height profile of trace gases.

There are several methods of mathematical calculation in the retrieval method; in this study, the Rodgers optimal estimation method was used[7][8]. Since this method uses information concerning the line shape of the observed spectrum, the frequencies are finely divided to create vector **y** with m components y_1, y_2, ..., y_m, representing the spectral intensities at each frequency. Next, the height profile of the target trace gas is defined by vector **x** with n components x_1, x_2, ..., x_n, representing the abundance of the gas at each altitude level. If the weighting function is defined as **K** (= **y**/**x**), then the relationship between the observed spectrum and the height profile of the trace gas can be expressed as **y** = **Kx**. In the Rodgers optimal estimation method, by assigning the observed spectrum **y**, a prior information of the height profile $\mathbf{x}_a$ (a priori), and their covariance matrix $\mathbf{S}_y$ and $\mathbf{S}_a$, respectively, the most likely height profile $\hat{\mathbf{x}}$ can be derived from the following equation.

$$\hat{\mathbf{x}}=\mathbf{x}_a+\mathbf{S}_a\mathbf{K}^T(\mathbf{K}\mathbf{S}_a\mathbf{K}^T+\mathbf{S}_y)^{-1}(\mathbf{y}-\mathbf{K}\mathbf{x}_a)$$
$$=\mathbf{x}_a+\mathbf{D}(\mathbf{y}-\mathbf{K}\mathbf{x}_a) \qquad (1)$$

Here,

$$\mathbf{D}=\mathbf{S}_a\mathbf{K}^T(\mathbf{K}\mathbf{S}_a\mathbf{K}^T+\mathbf{S}_y)^{-1} \qquad (2)$$

For this report we have attempted to determine the height profile and total column amount of ozone based on the absorption spectrum in the frequency region of 3051.20 to 3051.90 cm^{-1} (the second overtone transition of the asymmetric stretching vibration of ozone; with a rotational quantum number J = 7 - 31) observed by the Poker Flat FTIR. The data were processed using the SFIT2 program developed jointly by NIWA and NASA Langley[4][5]. The observed spectrum was divided by frequency every 0.0035 cm^{-1} to create vector **y**. Fig.2 is an example of the absorption spectrum for ozone used in the analysis observed at Poker Flat at 9 AM (Alaska standard time) on March 23, 2001. In the bottom figure, crosses represent observed values and solid lines show values calculated by the retrieval method. The top figure shows the residuals for the observed values for the spectrum subtracted by the calculated values. The ozone transition in this frequency region was selected due to the small temperature dependence of the spectral intensity and for the small degree of overlap with the absorption lines of other molecules.

The height profile was sectioned into 29 layers for altitudes between 0 and 100 km to create vector **x**. US standard atmosphere 1976 [9] was used as a priori $\mathbf{x}_a$. Diagonal matrices were assigned for the covariance matrices $\mathbf{S}_a$ and $\mathbf{S}_y$ for a priori $\mathbf{x}_a$ and observed spectrum y, respectively. $\mathbf{S}_a$ was created assuming that uncertainty of a priori is 10% of it, and $\mathbf{S}_y$ was created assuming a spectral signal-to-noise ratio of 500. The SFIT2 requires information on the temperature and pressure information at each altitude during the spectrum acquisition to calculate the weighting function **K**. The daily temperature and pressure data near Poker Flat provided by the United Kingdom Meteorological Office (UKMO) were used for the available altitudes (0 to 50 km)[10]. The monthly mean temperature and pressure data for the area of 65 °N provided by CIRA86 was

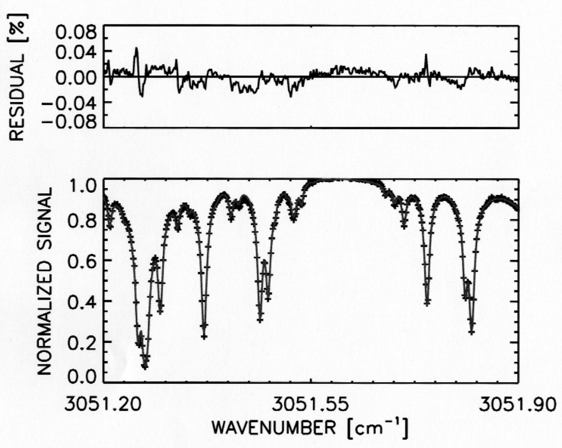

Fig.2 (Bottom) The infrared absorption spectrum for ozone observed at Poker Flat at 9 AM (Alaska standard time) on March 23, 2001. Crosses represent observed values and solid lines show values calculated by the retrieval method
(Top) The residual for the observed values for the spectrum subtracted by the calculated values

used with respect to altitudes for which UKMO data were not provided (50 to 100 km) [11]. Spectroscopic parameters, such as center frequencies, absorption intensities, and pressure-broadening coefficients of absorption spectra were obtained from the 1996 HITRAN database [12].

In the analysis using the retrieval method, **A** = **DK** is referred to as the averaging kernel, which is an index indicating the amount of information for the height profile that can be obtained from the spectrum. Fig.3 is an example of an averaging kernel obtained from analysis of the absorption spectrum for ozone (frequency region of 3051.20 to 3051.90 cm^{-1}) observed at Poker Flat at 9 AM (Alaska standard time) on March 23, 2001. The region with a large peak in the averaging kernel (at an altitude of 10 to 25 km) corresponds to the altitude in respect of which the most information may be obtained for the height profile based on the absorption spectrum. The peak of the averaging kernel gradually increases at altitudes higher than 10 km, because although the volume mixing ratio of ozone is small in the troposphere, it increases with altitude in the stratosphere, and so the abundance per unit volume (number density) also increases,

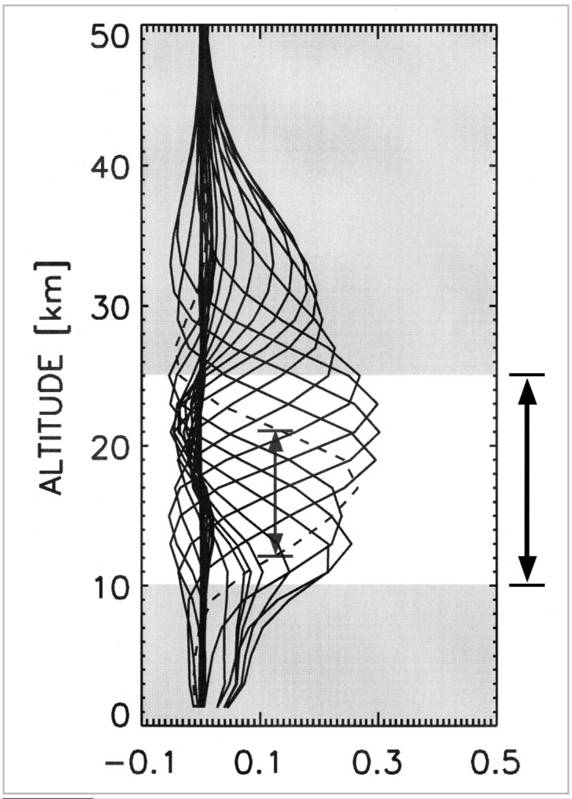

Fig.3 The averaging kernel obtained from analysis of the infrared absorption spectrum for ozone (3051.20 to 3051.90 cm^{-1}) observed at Poker Flat at 9 AM (Alaska standard time) on March 23, 2001

The region with the peaks of the averaging kernel (double-sided arrow to the right: altitude of 10 to 25 km) represents the altitude range where the largest amount of information on the height profile can be obtained from the absorption spectrum. The full width at half maximum of the averaging kernel (double-sided arrow inside plot: approximately 10 km) shows the height resolution of the obtained height profile.

resulting in a greater amount of available information for the height profile. The peak of the averaging kernel decreases above altitudes of 25 km because the ozone number density decreases along with the exponential decrease in atmospheric number density with altitude, resulting in less available information relating to the height profile. The height resolution of the obtained height profile can be approximated by the full width at half maximum of the averaging kernel (approximately 10 km at an altitude of 10 to 25 km). The large full width at half maximum of the averaging kernel above 25 km corresponds to the decrease in height resolution of the height profile because the Doppler broadening surpasses the pressure broadening and dominates the line shape of the absorption spectrum.

4 Results

Fig.4 is an example of the ozone height profile obtained using the retrieval method for an observation at Poker Flat at 9 AM (Alaska standard time) on March 23, 2001. Dashed and solid lines represent a priori and the retrieved height profile determined by the retrieval method, respectively. For comparison with the height profile obtained by FTIR observation, the results of observation by balloons (ozonesonde) launched from Fairbanks (64.81 °N, 147.86 °W; approximately 40 km southwest of Poker Flat) at 10 AM on the same day are shown by a dotted line. In the altitude range of 10 to 25 km, where abundant height-profile information is believed to be available based on the spectrum, the results of analysis using the retrieval method coincide well with the observed values for ozonesonde at altitude of 17 to 25 km, whereas the FTIR values are reduced to 1/2 to 1/3 of those obtained by ozonesonde at altitude of 10 to 17 km.

The height profile and the total column amount of ozone were determined using the retrieval method based on the ozone absorption spectra observed during the period from February 24 to October 28, 2001 under the conditions described above. Fig.5 shows the total column amount of ozone at Poker Flat in 2001. The circles in the figure represent the total column amount calculated using the retrieval method. For comparison, the solid line indicates observation data measured by TOMS (Total Ozone Mapping Spectrometer) on the Earth Probe satellite (Earth Probe/TOMS)[13] in 2001 near Fairbanks (i.e., a region within a radius of 40 km from Fairbanks). It can be seen that the values and trends observed using FTIR are generally consistent with Earth Probe/TOMS observations in terms of upward/downward trends on time scale of several tens of days from the end of April to May and the gradual downward/upward trends from June to October.

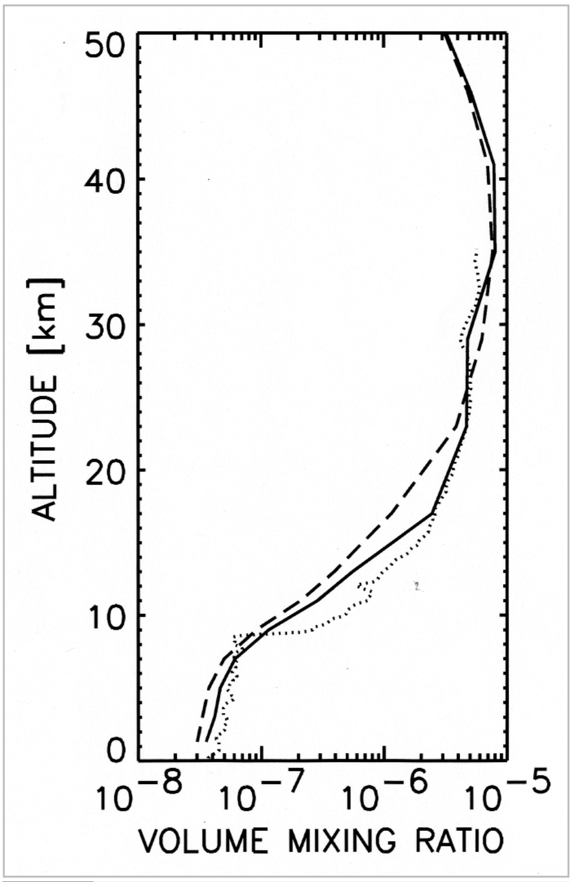

Fig.4 *The ozone height profile observed at Poker Flat at 9 AM (Alaska standard time) on March 23, 2001*

Dashed and solid lines represent a priori and the retrieved height profile determined by the retrieval method, respectively. The dotted line shows values observed by balloons (ozonesonde) launched from Fairbanks (approximately 40 km southwest of Poker Flat) at 10 AM on the same day.

5 Discussions

Height profiles were obtained based on the fact that the line shape of the ozone absorption spectrum at 3051.20 to 3051.90 cm^{-1} observed by FTIR is determined by altitude-dependent (i.e., pressure-dependent) pressure broadening. As stated previously in Section 3, since abundance of ozone is small at altitudes below 10 km and above 25 km, and the Doppler broadening dominates the line shape of the absorption spectrum above 25 km, little information is available for the height profile at the altitudes. Comparison of the FTIR ozone-height

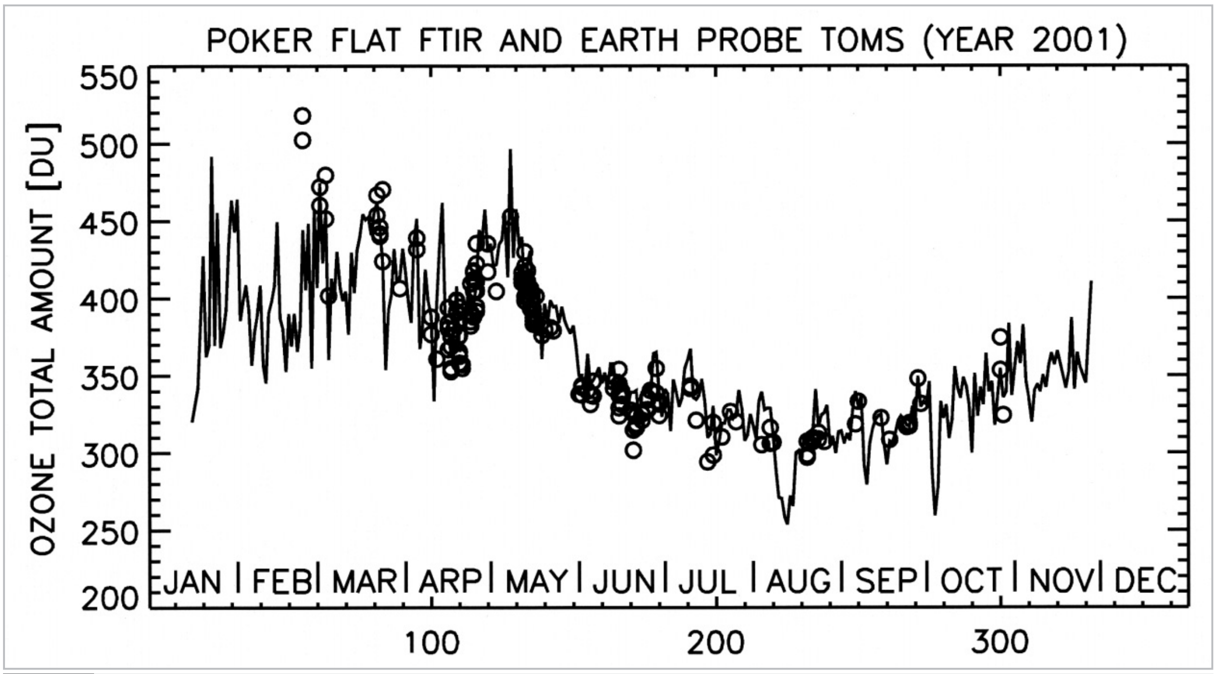

Fig.5 *The total column amount of ozone at Poker Flat determined by the retrieval method*

All data collected from FTIR observation for the 73-day period from February 24 to October 28, 2001 are indicated with circles. The solid line represents observation data collected by TOMS (Total Ozone Mapping Spectrometer) on the Earth Probe satellite in 2001 near Fairbanks (for the region within a radius of 40 km from Fairbanks).

profile (Fig.4) and ozonesonde observation in the altitude range of 10 to 25 km (a region for which information was available) shows a fair degree of consistency at altitudes of 17 to 25 km. On the other hand, the FTIR values are approximately 1/2 to 1/3 of those observed by ozonesonde at altitudes of 10 to 17 km. Several factors may contribute to such discrepancies. One is that the ozonesonde observation measures the local ozone volume mixing ratio at height resolutions of approximately 0.1 km, while the FTIR observes the ozone volume mixing ratio at height resolutions of 10 km, which may result in the observation of different regions in the vertical direction. A second possibility may lie in the fact that ozonesonde measurements are taken while adrift on the wind, while FTIR observations are always made in the direction of the sun; thus the observed regions for FTIR and ozonesonde may be separated by several tens of kilometers in the horizontal direction. It may also be possible that the 1-hour difference in the observation time for FTIR and ozonesonde resulted in the observations of the temporal and spatial differences in the ozone mixing ratio. Another contributing factor to the discrepancies may lie in the lack of consideration of the non-linear term for trace-gas abundance in the radiative transfer model of the atmosphere used in the SFIT2 program, resulting in underestimation of the ozone mixing ratio for FTIR.

Although not shown in Fig.5, in terms of the long-term trends in total ozone variation for the period from June to September, the ratio between FTIR and TOMS values indicated that the estimated FTIR values tended to be several percent lower than TOMS. This may result from the use (under the retrieval method) of a single ozone profile as a priori throughout the year; this approach would not take actual seasonal ozone variations into account and may have resulted in systematic seasonal errors. Such effects may be removed by using seasonal profiles as a priori.

6 Conclusions

As part of the international collaborative project (Alaska Project) between the Communications Research Laboratory (CRL) and the University of Alaska Fairbanks (UAF), measurements of infrared solar absorption spectra have been performed since July 1999 using the Fourier transform infrared spectrometer (FTIR) at the UAF's Poker Flat Research Range, with the aim of conducting observations of atmospheric trace gases in the troposphere and the stratosphere at the polar region. Conventional studies consisted mainly of a determination of the total column amount of trace gases based on the total intensity of their absorption spectra using the SFIT program. However, height profiles of trace gases cannot be obtained with SFIT, nor can precise information on the total column amount be determined. Thus, beginning around 1995, numerous studies have focused on determining the height profiles of trace gases from the line shape of their absorption spectra using the SFIT2 program. However, few such studies using the retrieval method were conducted in Japan before 2001. At present CRL is promoting the introduction of SFIT2, with the cooperation of the National Institute for Environmental Studies and Tohoku University, and is currently making adjustments to analysis conditions and attempting to obtain a height profile.

In this paper, we discussed the determination of the daily ozone height profile using the retrieval method and based on the ozone absorption spectra observed during the nine-month period from February to October 2001. The results were compared with data obtained from the ozonesonde and the Earth Probe/TOMS for purposes of validation. For the altitude range of 10 to 25 km, where abundant information can be obtained from the ozone absorption spectra, the ozone height profiles observed by FTIR coincided well with the ozonesonde data above 17 km, but were reduced to 1/2 to 1/3 of the ozonesonde values in the 10 to 17 km range. This discrepancy is most likely due to differences in height resolutions or in the atmospheric regions observed by FTIR and the ozonesonde, or to terms not considered in the atmospheric radiative transfer model.

The results of FTIR and Earth Probe/TOMS observations concerning the total column amount of ozone were generally consistent during the period from February to October 2001, and variations on time scales of longer than 1 to 2 months were also well reproduced. However, the values estimated for FTIR were found to be several percent lower during the months of June to September. The use of a single ozone profile as a priori throughout the year may be the cause of erroneous seasonal variations.

In the future, we plan to determine the abundance of other trace gases, including those related to global warming (such as carbon dioxide and methane), and to discuss daily, seasonal, and year-to-year variations in trace gases using abundant observation data. In the Alaska Project, observations with a millimeter-wave radiometer are also being conducted at the Poker Flat Research Range, in an effort to determine the abundance of atmospheric trace gases from the stratosphere to the mesosphere (i.e., within an altitude range of 20 to 60 km). Simultaneous observations with the FTIR and the millimeter-wave radiometer should provide information for a wide range of altitudes (from the troposphere to the mesosphere) in terms of common subjects of observation such as ozone. Therefore, joint studies using the FTIR and the millimeter-wave radiometer are planned.

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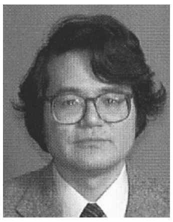
Observational Study of Middle Atmosphere



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