

## Theoretical Evaluation of Earth-to-Satellite Laser Long-Path Absorption Measurement of Atmospheric Trace Species in the Infrared Region

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Measurement of atmospheric trace species based on the earth-to-satellite laser long-path absorption method using continuously tunable infrared laser was theoretically studied. Optimum wavelengths for the measurement were systematically investigated in the infrared region for all molecules included in the HITRAN92 data base. It was shown that H<sub>2</sub>O, CO<sub>2</sub>, O<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O and CO have strong absorption lines, and that the inversion method for retrieving vertical profiles can be applied. Also, it was shown that absorption lines of NO<sub>2</sub>, OCS, HCl, C<sub>2</sub>H<sub>6</sub>, HF, HNO<sub>3</sub>, NO, NH<sub>3</sub>, C<sub>2</sub>H<sub>2</sub>, H<sub>2</sub>CO, and HOCl have sufficient intensities for measuring column contents. Accuracy of column measurement of each molecule was estimated for a laser long-path absorption system using a geosynchronous satellite.

**KEYWORDS:** laser long-path absorption method, satellite remote sensing, atmospheric trace species, tunable infrared laser

### 1. Introduction

The earth-to-satellite laser long-path absorption method is one of the most sensitive remote sensing techniques for measuring atmospheric trace species. In tropospheric measurement, where passive satellite sensors are not necessarily useful, this technique can play a unique role.

There are two methods for measurement based on earth-to-satellite laser long-path absorption. One is the method using a retroreflector on a satellite, and the other is that using a detection system on a satellite. The concept of measurement using a satellite retroreflector was discussed by Hinkley.<sup>1)</sup> In the method, a laser beam is transmitted from a ground station, reflected by the satellite retroreflector, and received at the ground station. Absorption of the atmosphere is measured in the round-trip path. The first experiment based on this method is planned by our group with a cube-corner retroreflector called RIS<sup>2-5)</sup> installed on a Japanese polar-orbit satellite known as ADEOS, which is scheduled for launch in 1996.

The method using a detection system on a geosynchronous satellite was studied by Sugimoto.<sup>6)</sup> In this method, a laser beam transmitted from a ground station is received with a detection system on a geosynchronous satellite. The received signal is transferred to the ground. In this method, the ground station can be greatly simplified compared with the method using a satellite retroreflector, because it does not require a large receiving telescope and a wide-range tracking system. Also, the system can be designed for continuous measurements from multiple ground stations.

In this paper, we report on evaluation of the earth-to-satellite laser long-path absorption method from a spectroscopic point of view. We considered a system consisting of a geosynchronous satellite and a ground station with a continuously tunable infrared laser. We investigated absorption lines of trace species systematically in the infrared region from 1 to 20  $\mu\text{m}$  using the HITRAN92 data base and the FASCOD program for calculation of synthesized absorption spectrum. To

evaluate the column measurement of trace species with weak absorption lines, we developed an algorithm for estimating column density and error in the measurement. We applied this method to determine the optimum wavelength region for measurement of each species. Also, we estimated the accuracy of the column density measurement.

### 2. Earth-to-Satellite Laser Long-Path Absorption System

Figure 1 shows the concept of earth-to-satellite laser long-path absorption measurement using a detection system on a geosynchronous satellite. A laser beam

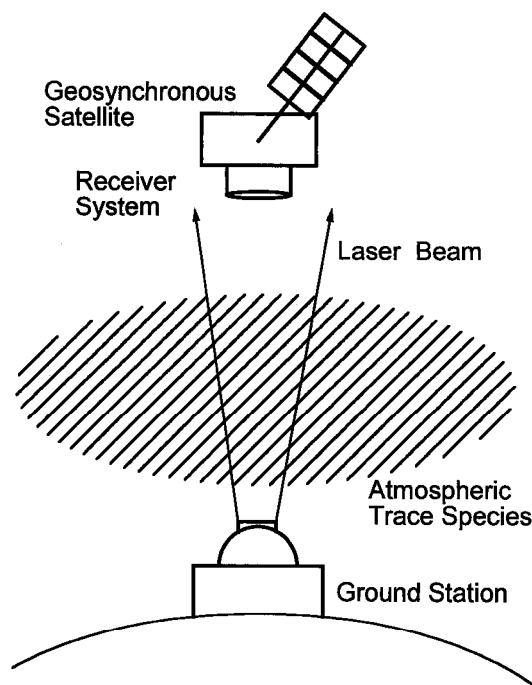


Fig. 1. Concept of earth-to-satellite laser long-path absorption measurement using a detection system on a satellite.

transmitted from a ground station is received by a detection system on the satellite. The absorption spectrum of atmospheric trace species is measured by scanning the laser wavelength. Concentration of the target species is obtained from the spectrum. For species having sufficiently strong absorption lines, we can apply the inversion method which utilizes pressure dependence of absorption line shape to retrieve the vertical profile of the species. For species with weak absorption lines, we can obtain column content by the least squares method.

We considered a system with the parameters listed in Table I and evaluated the signal-to-noise ratio (SNR) of optical detection in the system. The signal photoelectron number received at the satellite is written by

$$n = E_0 \eta T(\nu) D^2 / (R \theta_t)^2 / (h\nu), \quad (1)$$

where  $E_0$ ,  $\eta$ ,  $T(\nu)$ ,  $D$ ,  $R$ , and  $\theta_t$  are the parameters defined in Table I, and  $h\nu$  represents the energy of the photon. From eq. (1), the number of received photoelectrons at  $10 \mu\text{m}$  is estimated at  $7.0 \times 10^5$  for the system with the parameters in Table I. Laser energy required for the measurement depends on atmospheric transmittance at the absorption line of the target species. We considered the worst transmittance of 0.01 here and a laser pulse energy of 2 mJ.

There are two major noise sources in the measurement. One is the detector, and the other is the radiation from the ground. Detector noise can be estimated by detectivity,  $D^*$ , which is written as

$$D^* = A_d^{1/2} / \text{NEP}, \quad (2)$$

where  $A_d$  is area of the detector and NEP is noise equivalent power. Using eq. (2), we can calculate the noise equivalent photoelectron number,  $N_d$ , by

$$N_d = [A_d \tau]^{1/2} / [D^* h\nu], \quad (3)$$

where  $\tau$  is the time constant of detection. The typical value of  $D^*$  for infrared detectors such as HgCdTe and InSb is approximately  $2 \times 10^{10} \text{ cm Hz}^{1/2} \text{ W}^{-1}$ ,<sup>7)</sup> and then  $N_d$  can be estimated at  $4.9 \times 10^3$ .

Background radiation from the ground is calculated by

$$N_r = R_e (\pi D^2 / 4R^2) (\pi \theta_t^2 R^2 / 4) \eta \Delta\mu \tau / (h\nu) \\ + a R_{\text{sun}} (\pi \theta_t^2 R^2 / 4) \{ (\pi D^2 / 4) / (2\pi R^2) \} \eta \Delta\mu \tau / (h\nu). \quad (4)$$

The first term represents infrared radiation from the ground and the second term represents the reflected solar radiation.  $R_e$  is spectral radiance of the earth, and  $R_{\text{sun}}$  is the solar spectral irradiance.  $a$  in the second term represents albedo of the earth. We assumed  $a = 1$ .  $R_e$  depends on season and latitude, and the maximum value is approximately  $10 \text{ Wm}^{-2} \text{ sr}^{-1} \mu\text{m}^{-2}$  at approximately  $9 \mu\text{m}$ .<sup>8)</sup>  $R_{\text{sun}}$  is less than  $1 \text{ Wm}^{-2} \mu\text{m}^{-1}$  at wavelengths longer than  $5 \mu\text{m}$ .<sup>9)</sup> By using the parameters listed in Table I, the first and second terms in eq. (4) are estimated at  $1.6 \times 10^4$  and 77, respectively. Noise due to background radiation,  $N_d$ , is expressed by the square root of  $N_r$ , and is approximately 127 photoelectrons.

Table I. Parameters of earth-to-satellite laser long-path absorption system.

Altitude of satellite: $R$	36,000 km
Transmitted laser power: $E_0$	2 mJ
Laser wave number: $\nu$	2–14 $\mu\text{m}$
Beam divergence of transmitter: $\theta_t$	50 $\mu\text{rad}$
Diameter of receiver telescope: $D$	0.15 m
Field of view of receiver: $\theta_r$	0.278 mrad (10 km foot print)
Spectral bandwidth of receiver: $\Delta\mu$	3 $\mu\text{m}$
Time constant of detector: $\tau$	100 ns
Area of detector: $A_d$	$3.79 \times 10^{-5} \text{ cm}^2$
Detectivity: $D^*$	$2.0 \times 10^{10} \text{ cmHz}^{1/2} \text{ W}^{-1}$
Overall efficiency of detection: $\eta$	0.1
Spectral radiance from the earth: $R_e$	$10 \text{ Wm}^{-2} \text{ sr}^{-1} \mu\text{m}^{-1}$ (maximum at 9 $\mu\text{m}$ )
Solar spectral radiance: $R_{\text{sun}}$	$1 \text{ Wm}^{-2} \mu\text{m}^{-1}$ (typical at > 5 $\mu\text{m}$ )
Albedo of the earth: $a$	0.3
Atmospheric transmittance: $T(\nu)$	0.01 (minimum)

From the estimation described above, it can be seen that the dominant noise in the measurement is detector noise. It can also be concluded that theoretical SNR of detection of a laser pulse is larger than 140 with the parameters in Table I. In addition to the noise considered above, we may have to consider the effect of atmospheric turbulence in actual measurement, though the effect can be canceled, in principle, by using a reference laser which is transmitted simultaneously with the laser for the spectrum measurement. It is, however, difficult to estimate theoretically the remaining error due to turbulence when a reference laser is used. Consequently, we assumed in the following estimation that we can achieve SNR = 100 for detection of a laser pulse including turbulence effect.

### 3. Survey of Strongest Absorption Lines

To study the feasibility of the earth-to-satellite laser long-path absorption method, we surveyed absorption lines of trace species systematically using HITRAN92<sup>10)</sup> and the FASCOD.<sup>11)</sup> First, a data base of high resolution atmospheric absorption spectra was constructed for looking up the transmittance at absorption lines of target molecules. The absorption spectrum was calculated with FASCOD for the vertical path in the wavelength range from 1–20  $\mu\text{m}$  with a spectral resolution of  $0.001 \text{ cm}^{-1}$ . All molecules were included, and the midlatitude summer model was used for vertical profiles in the calculation with FASCOD.

On the other hand, we searched for the strongest absorption line for each species among all the lines compiled in HITRAN92. Table II lists the strongest absorption lines of species included in HITRAN92. Absorption of the molecules and atmospheric transmittance at the wavelengths of the absorption lines, which were calculated with FASCOD, are also listed in Table II.

Molecules from  $\text{H}_2\text{O}$  to CO in Table II have strong absorption, and there are many absorption lines suitable for applying the inversion method to retrieve vertical profiles. In this study, however, we focused on the

feasibility of measurement of column contents of molecules having weak absorption, because these trace species are important in atmospheric chemistry. Therefore, we developed an algorithm for estimating error in the column measurement and estimated error in the following way. First, we select the strongest absorption line. If the transmittance at the selected line is smaller than  $1.0 \times 10^{-2}$ , we select the next strongest line.

#### 4. Measurement of Column Content

We assumed that the spectral bandwidth of the laser is sufficiently small compared to the width of the absorption line of the target molecule. Received signal,  $E_i$ , can be written as

$$E_i = \eta E_0 T(\nu_i) + \varepsilon_i, \quad (5)$$

where  $i$  represents data point in a wavelength scan.  $\eta$ ,  $E_0$ ,  $T(\nu_i)$ , and  $\varepsilon_i$  represent a system constant, laser pulse energy, transmittance at the wave number  $\nu_i$ , and noise in the signal, respectively.

Noise dominant in the signal is detector noise and background noise. These noises are independent of the signal intensity. Therefore, variance of  $\varepsilon_i$  is constant. Also, we assumed that covariance of  $\varepsilon_i$  at different wavelengths is zero.

$$\begin{aligned} \langle \varepsilon_i \varepsilon_j \rangle &= \varepsilon_0^2 \quad (i=j) \\ &0 \quad (i \neq j) \end{aligned} \quad (6)$$

Column transmittance of the atmosphere,  $T(\nu_i)$ , can be written by

$$T(\nu_i) = \exp(-q_1 \sigma_1(\nu_i) - q_2 \sigma_2(\nu_i) - \alpha_0), \quad (7)$$

where  $q_1$  and  $q_2$  represent column contents of target molecule and interfering molecule, respectively.  $\sigma_1(\nu_i)$  and  $\sigma_2(\nu_i)$  are effective column absorption cross sections of target molecule and interfering molecule, respectively. We considered the single interfering molecule in eq. (7), for simplicity.  $\alpha_0$  is continuous absorption due to water vapor and aerosols. We assumed that  $\alpha_0$  is constant within the wavelength range of the scan.

To apply the linear least squares method, we rewrite eq. (5) by taking the logarithm.

$$\begin{aligned} y_i &= -\ln E_i \\ &= -\ln \eta - \ln E_0 - \ln T(\nu_i) - \varepsilon_i / \{E_0 T(\nu_i)\}. \end{aligned} \quad (8)$$

The logarithm term including  $\varepsilon_i$  was expanded, and higher-order terms are neglected in eq. (8). By substituting eq. (7) into eq. (8), we obtain

$$\begin{aligned} y_i &= -\ln \eta - \ln E_0 + q_1 \sigma_1(\nu_i) + q_2 \sigma_2(\nu_i) \\ &\quad + \alpha_0 - \varepsilon_i / \{\eta E_0 T(\nu_i)\}. \end{aligned} \quad (9)$$

Consequently,  $y_i$  is written as a linear transformation of three unknown parameters  $x_1$ ,  $x_2$ , and  $x_3$ .

$$y_i = x_1 q_{10} \sigma_1(\lambda_i) + x_2 q_{20} \sigma_2(\lambda_i) + x_3 - \varepsilon_i / \{K P_0 T(\lambda_i)\}, \quad (10)$$

where  $x_1$ ,  $x_2$ , and  $x_3$  are defined by

$$\begin{aligned} x_1 &= q_1 / q_{10}, \\ x_2 &= q_2 / q_{20}, \\ x_3 &= -\ln \eta - \ln E_0 + \alpha_0. \end{aligned} \quad (11)$$

Table II. Strongest absorption lines of atmospheric molecules.

Molecule	Wave number (cm <sup>-1</sup> )	Absorption <sup>a)</sup>	Atmospheric transmittance <sup>b)</sup>
H <sub>2</sub> O	1684.835200	1.000000E+00	
CO <sub>2</sub>	2361.466037	1.000000E+00	
O <sub>3</sub>	1052.848400	1.000000E+00	
CH <sub>4</sub>	3067.300100	1.000000E+00	
N <sub>2</sub> O	2236.223500	1.000000E+00	1.765983E-36
CO	2172.758811	9.999981E-01	1.345158E-06
NO <sub>2</sub>	1628.836000	1.400772E-01	5.527349E-37
OCS	2070.510000	1.229653E-01	3.825258E-01
HCl	2944.913771	8.785880E-02	3.229543E-01
C <sub>2</sub> H <sub>6</sub>	2986.723000	7.947700E-02	3.316685E-01
HF	4038.962500	4.085960E-02	4.077121E-01
HNO <sub>3</sub>	1325.960000	3.810060E-02	2.612591E-03
NO	1903.123100	3.529210E-02	4.921505E-03
NH <sub>3</sub>	967.346200	1.679420E-02	7.825342E-01
C <sub>2</sub> H <sub>2</sub>	729.559100	1.326180E-02	1.129241E-02
HCN	3337.139200	1.302840E-02	1.860056E-08
H <sub>2</sub> CO	2814.743400	4.832700E-03	6.620913E-01
HOCl	1253.184600	4.668000E-04	2.961914E-01
SO <sub>2</sub>	1373.078560	0.000000E+00	
OH	3568.416700	0.000000E+00	
HBr	2620.997515	0.000000E+00	
HI	2288.609701	0.000000E+00	
ClO	859.768200	0.000000E+00	
CH <sub>3</sub> Cl	2965.777300	0.000000E+00	
H <sub>2</sub> O <sub>2</sub>	1284.204500	0.000000E+00	
PH <sub>3</sub>	991.958000	0.000000E+00	
COF <sub>2</sub>	1953.377500	0.000000E+00	
SF <sub>6</sub>	949.733700	0.000000E+00	
H <sub>2</sub> S	1292.863040	0.000000E+00	

<sup>a)</sup>Absorption, (1-transmittance), of target molecule in earth-to-satellite path with 90° elevation angle.

<sup>b)</sup>Transmittance of the atmosphere including all molecules in the same optical path.

$x_1$  and  $x_2$  are the normalized column contents which are normalized by standard values,  $q_{10}$  and  $q_{20}$ .

Equation (10) can be written in a matrix form by

$$\mathbf{Y} = \mathbf{A} \mathbf{X}. \quad (12)$$

$$\mathbf{Y} = \begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_n \end{pmatrix}$$

$$\mathbf{A} = \begin{pmatrix} q_{10} \sigma_1(\nu_1) & q_{20} \sigma_2(\nu_1) & 1 \\ q_{10} \sigma_1(\nu_2) & q_{20} \sigma_2(\nu_2) & 1 \\ \vdots & \vdots & \vdots \\ q_{10} \sigma_1(\nu_n) & q_{20} \sigma_2(\nu_n) & 1 \end{pmatrix}$$

$$\mathbf{X} = \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix}$$

Vector  $\mathbf{X}$  determined by the least squares method can be expressed by

$$\mathbf{X}_{\text{est}} = (\mathbf{A}^T \Sigma_y \mathbf{A})^{-1} \mathbf{A}^T \Sigma_y \mathbf{Y}, \quad (13)$$

where  $\Sigma_y$  represents covariance matrix of  $\mathbf{Y}$ .

$$\begin{aligned}
 (\Sigma_y)_{ij} &= \langle \{ \epsilon_i / \{ \eta E_0 T(\nu_i) \} \} [ \epsilon_j / \{ \eta E_0 T(\nu_j) \} ] \rangle^{-1} \\
 &= \{ \epsilon_0 / \eta E_0 T(\nu_i) \}^{-2} \quad (i=j) \\
 &0 \quad (i \neq j)
 \end{aligned}
 \tag{14}$$

**5. Optimization of Wavelength Range for Scan**

To determine optimum wavelength range for spectrum measurement, we derived a function which describes error in the obtained column content. Variance of parameter  $x_i$  which is determined by the least squares method is expressed by a diagonal element of the covariance matrix,  $\Sigma_x$ , which is defined by

$$\Sigma_x = (\mathbf{A}^T \Sigma_y \mathbf{A})^{-1}
 \tag{15}$$

Therefore, variance of determined column content is estimated by

$$\sigma^2 x_1 = (\Sigma_x)_{11}
 \tag{16}$$

Elements of matrix  $\mathbf{A}^T \Sigma_y \mathbf{A}$  are written by

$$(\mathbf{A}^T \Sigma_y \mathbf{A})_{11} = \epsilon^{-2} \Sigma \sigma_1(\nu_i)^2 T(\nu_i)^2 = \epsilon^{-2} a / \Delta \nu,
 \tag{17 \cdot a}$$

$$(\mathbf{A}^T \Sigma_y \mathbf{A})_{22} = \epsilon^{-2} \Sigma \sigma_2(\nu_i)^2 T(\nu_i)^2 = \epsilon^{-2} b / \Delta \nu,
 \tag{17 \cdot b}$$

$$(\mathbf{A}^T \Sigma_y \mathbf{A})_{33} = \epsilon^{-2} \Sigma T(\nu_i)^2 = \epsilon^{-2} \Sigma T(\nu_i)^2 c / \Delta \nu,
 \tag{17 \cdot c}$$

$$\begin{aligned}
 (\mathbf{A}^T \Sigma_y \mathbf{A})_{23} &= (\mathbf{A}^T \Sigma_y \mathbf{A})_{32} \\
 &= \epsilon^{-2} \Sigma \sigma_2(\nu_i) T(\nu_i)^2 = \epsilon^{-2} \alpha / \Delta \nu,
 \end{aligned}
 \tag{17 \cdot d}$$

$$\begin{aligned}
 (\mathbf{A}^T \Sigma_y \mathbf{A})_{13} &= (\mathbf{A}^T \Sigma_y \mathbf{A})_{31} \\
 &= \epsilon^{-2} \Sigma \sigma_1(\nu_i) T(\nu_i)^2 = \epsilon^{-2} \beta / \Delta \nu,
 \end{aligned}
 \tag{17 \cdot e}$$

$$\begin{aligned}
 (\mathbf{A}^T \Sigma_y \mathbf{A})_{12} &= (\mathbf{A}^T \Sigma_y \mathbf{A})_{21} \\
 &= \epsilon^{-2} \Sigma \sigma_1(\nu_i) \sigma_2(\nu_i) T(\nu_i)^2 = \epsilon^{-2} \gamma / \Delta \nu,
 \end{aligned}
 \tag{17 \cdot f}$$

where

$$\epsilon = \epsilon_0 / (\eta E_0)
 \tag{18}$$

Also,  $a, b, c, \alpha, \beta,$  and  $\gamma$  are defined by

$$a = \int \sigma_1(\nu_i)^2 T(\nu_i)^2 d\nu,
 \tag{19 \cdot a}$$

$$b = \int \sigma_2(\nu_i)^2 T(\nu_i)^2 d\nu,
 \tag{19 \cdot b}$$

$$c = \int T(\nu_i)^2 d\nu,
 \tag{19 \cdot c}$$

$$\alpha = \int \sigma_2(\nu_i) T(\nu_i)^2 d\nu,
 \tag{19 \cdot d}$$

$$\beta = \int \sigma_1(\nu_i) T(\nu_i)^2 d\nu,
 \tag{19 \cdot e}$$

$$\gamma = \int \sigma_1(\nu_i) \sigma_2(\nu_i) T(\nu_i)^2 d\nu,
 \tag{19 \cdot f}$$

From eq. (17), variance of the determined column content can be written by

$$\begin{aligned}
 \sigma^2 x_1 &= (\Sigma_x)_{11} = ((\mathbf{A}^T \Sigma_y \mathbf{A})^{-1})_{11} \\
 &= \epsilon^2 \Delta \nu (bc - \alpha^2) / (abc + 2\alpha\beta\gamma - a\alpha^2 - b\beta^2 - c\gamma^2).
 \end{aligned}
 \tag{20}$$

Because the time required for a scan measurement depends on the number of steps, we have to normalize the

variance to compare error in the same measurement time. The measurement time is proportional to the number of the steps which is written by

$$m = (\nu_m - \nu_1) / \Delta \nu,
 \tag{21}$$

where  $\nu_1$  and  $\nu_m$  indicate the minimum and the maximum wave number of the scan range, respectively. We can define normalized variance by the following equation.

$$\begin{aligned}
 (\sigma^2 x_1)_n &= m (\Sigma_x)_{11} \\
 &= \epsilon^2 F(\nu_1, \nu_m),
 \end{aligned}
 \tag{22}$$

where function  $F$  is defined by

$$\begin{aligned}
 F(\nu_1, \nu_m) &= (\nu_m - \nu_1) (bc - \alpha^2) \\
 &/ (abc + 2\alpha\beta\gamma - a\alpha^2 - b\beta^2 - c\gamma^2).
 \end{aligned}
 \tag{23}$$

It is seen from eq. (22) that the optimum scan range, i.e.,  $\nu_1$  and  $\nu_m$ , can be determined by finding the minimum of  $F(\nu_1, \nu_m)$ .

An example of the absorption spectrum of the atmosphere is shown in Fig. 2, which includes absorption lines of NO. Figure 3 shows the contour map of  $F(\nu_1, \nu_m)$  for NO. It can be seen in Fig. 3 that  $F(\nu_1, \nu_m)$  has a minimum when  $\nu_1 = 1900.057 \text{ cm}^{-1}$  and  $\nu_m = 1900.096 \text{ cm}^{-1}$ .

Table III lists the optimum scan ranges and the minimum values of  $F(\nu_1, \nu_m)$ . Figure 4 shows the atmospheric absorption spectrum and the optimum wavelength regions.

**6. Estimation of Error in Column Content Measurement**

Error in column content for the optimum scan range is obtained by eq. (20).  $\epsilon$  in eq. (18) can be written using the signal-to-noise ratio (SNR) of the optical detection by

$$\epsilon^2 = T'^2 / \text{SNR}^2,
 \tag{24}$$

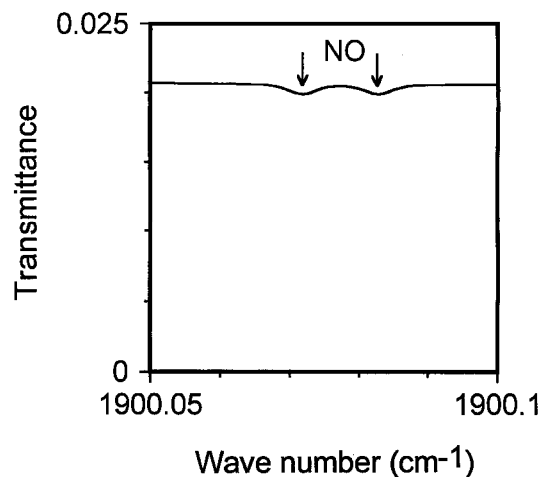


Fig. 2. Synthesized absorption spectrum of the atmosphere in the wavelength range including absorption lines of NO. Ground-to-satellite path, 90° elevation angle.

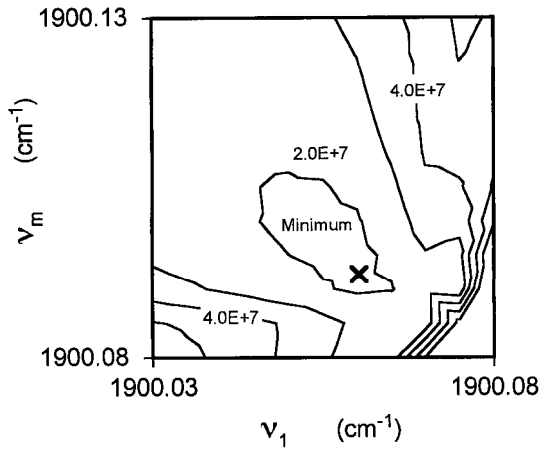


Fig. 3.  $F(\nu_1, \nu_m)$  for NO indicated by a contour map.  $\nu_1$  and  $\nu_m$  represent, respectively, the minimum and the maximum wave number of the scan range.

where  $T'$  represents averaged transmittance in the scan range. Consequently, error in the determined column density is written by

$$(\sigma^2 x_1)^{1/2} = \{F(\nu_1, \nu_m)/m\}^{1/2} T' / \text{SNR}. \quad (25)$$

From the result of discussion in §2, we assumed that SNR=100. Also, we assumed that the repetition rate of the laser is 10 Hz, measurement time is 1 min, and therefore  $m=600$ . The results of the estimation are listed in the fifth column in Table III. The results show that column contents of NO<sub>2</sub>, HNO<sub>3</sub>, HF, HCl, OCS, C<sub>2</sub>H<sub>6</sub>, NH<sub>3</sub>, NO, C<sub>2</sub>H<sub>2</sub>, H<sub>2</sub>CO HOCl can be measured with this method. The results also show that the measurements of HCN and molecules listed below HOCl in Table II are impossible with this method.

Table III. Optimum wavelength range, minimum  $F(\nu_1, \nu_m)$ , and estimated error in column content.

	$\nu_1(\text{cm}^{-1})$	$\nu_m(\text{cm}^{-1})$	$F(\nu_1, \nu_m)$	Error(%)	Transmittance	Laser energy (mJ) <sup>a)</sup>
NO	1900.057	1900.096	1.78E+07	3.69	0.021	1.0
NO <sub>2</sub>	2918.858	2918.889	2.18E+05	10.7	0.564	0.04
HNO <sub>3</sub>	1325.844	1325.895	8.69E+07	4.44	0.012	1.7
HF	4038.944	4038.983	2.48E+04	2.62	0.408	0.05
HCl	2944.900	2944.929	1.76E+04	1.72	0.318	0.06
OCS	2070.106	2070.171	4.92E+03	1.46	0.510	0.04
HCN	3337.124	3337.139	4.67E+15	—	—	—
C <sub>2</sub> H <sub>2</sub>	729.538	729.824	4.13E+08	25.7	0.031	0.65
C <sub>2</sub> H <sub>6</sub>	2986.543	2986.892	1.70E+04	1.85	0.348	0.06
NH <sub>3</sub>	967.189	967.387	6.53E+04	8.13	0.779	0.03
H <sub>2</sub> CO	2814.475	2814.771	1.91E+06	38.8	0.687	0.03
HOCl	1253.178	1253.194	2.09E+08	164.0	0.278	0.07

<sup>a)</sup>Laser pulse energy required for the measurement.

In the discussion of the measurement system in §2, we assumed that we use a laser with an output pulse energy of 2 mJ to obtain SNR=100 at the lowest atmospheric transmittance of 0.01. The required pulse energy, however, is inversely proportional to the transmittance. We list the required pulse energy for each molecule in the last column in Table III.

### 7. Discussion

The results of the evaluation show that a number of trace species which are important in the analysis of global atmospheric environment problems can be measured with the earth-to-satellite laser long-path absorption method. This method has a great advantage over passive techniques using solar radiation when the atmospheric transmittance at the absorption line of the target species is small. In the measurement of NO, for example, the transmittance at the target absorption

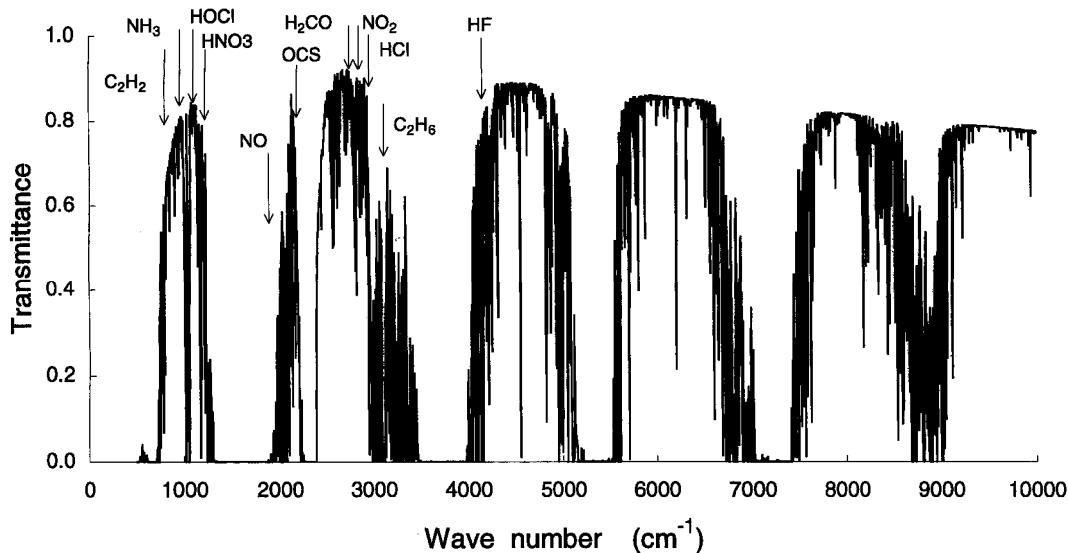


Fig. 4. Synthesized absorption spectrum of the atmosphere and the optimum wavelengths for column measurements of trace species.

line is very small due to absorption of water vapor, and it is almost impossible to measure the spectrum with passive techniques. With the laser long-path absorption method, however, column contents in the background atmosphere can be measured with sufficient accuracy.

The results of this study also show a limitation of the measurement method. It was shown that molecules with extremely weak absorption cannot be measured with this method. This limitation is common to all techniques based on open-air long-path absorption, including passive techniques.

The evaluation reported in this paper is based on the HITRAN92 data base, and therefore measurement of molecules which are not included in the data base was not evaluated. Also, the investigation was limited to the infrared region. However, the feasibility of measuring molecules such as CFCs in the infrared region and NO<sub>2</sub>, SO<sub>2</sub>, OH, etc., in the visible and near-ultraviolet regions must be studied further.

The inversion method to retrieve vertical profiles can be applied to H<sub>2</sub>O, CO<sub>2</sub>, O<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O and CO, because there are many absorption lines with various intensities. Further study will be required in order to optimize the wavelength range for the scan measurement for retrieving the vertical profile of each molecule.

There are several methods for generating tunable infrared laser pulses for the measurement. Optical parametric oscillator/amplifier<sup>12)</sup> and difference frequency mixing<sup>13)</sup> using crystals such as AgGaS<sub>2</sub>, AgGaSe<sub>2</sub>, and ZnGeP<sub>2</sub> are candidates. The pulse energy required for the measurement can be achieved with these methods for most of the molecules listed in Table III. An alternative approach is harmonics generation of a tunable single-mode high-pressure CO<sub>2</sub> laser.<sup>14)</sup> The wavelengths for molecules in Table III except for C<sub>2</sub>H<sub>2</sub> and HOCl can be covered with second and third harmonics of the

high-pressure CO<sub>2</sub> laser. Further study on the laser system is also required for constructing an earth-to-satellite laser long-path absorption system.

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