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Technical Memorandum TM 78067

Stratospheric Ozone Measurement With An Infrared Heterodyne Spectrometer

(NASA-TM-78067) STRATOSPHERIC OZONE MEASUREMENT WITH AN INFRARED HETERODYNE SPECTROMETER (NASA) 16 p HC A02/MF A01 CSCL 04A

N78-17532

Unclas G3/46 05050

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JANUARY 1978

National Aeronautics and Space Administration

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STRATOSPHERIC OZONE MEASUREMENT WITH AN INFRARED HETERODYNE SPECTROMETER

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ABSTRACT

A stratospheric ozone absorption line in the 10μm band has been measured and resolved completely, using an infrared heterodyne spectrometer with a spectral resolution of 5 MHz (0.000167 cm^{-1}). The vertical concentration profile of stratospheric ozone is obtained through an analytical inversion of the measured spectral line profile. The absolute total column density was 0.34 cm-atm with a peak mixing ratio occurring at $\sim$ 24 km. The (7,1,6) - (7,1,7) O_3 line center frequency was found to be 1043.1775 $\pm$ 0.00033 cm^{-1}, or 430 $\pm$ 10 MHz higher than the P(24) CO_2 laser line frequency.

STRATOSPHERIC OZONE MEASUREMENT WITH AN INFRARED HETERODYNE SPECTROMETER

This research note reports measurements of a stratospheric ozone concentration profile through detection of infrared absorption lines with a heterodyne spectrometer. These measurements were made on April 7, 1977 by observing the sun in the $9.4\mu m$ band with a spectral resolution of 5 MHz ($0.000167\ cm^{-1}$). This resolution, the highest employed so far for detection of stratospheric lines, is adequate to resolve the absorption lines completely, allowing an inversion of the lines for evaluation of vertical concentration profiles. Similar retrievals based on heterodyne measurements with lower spectral resolution [500 MHz or $0.0167\ cm^{-1}$ and 30 MHz or $0.001\ cm^{-1}$] have been reported previously (Menzies and Seals, 1977; Peyton et al. 1977). Diode laser heterodyne spectra of atmospheric O_3 at a resolution of 70 MHz have also been reported (Frerking and Muehlner, 1977) and altitude profiles have been derived (Abbas et al. 1977).

The heterodyne spectrometer employed in the present measurements is based on a line-by-line tunable CO_2 laser, a liquid nitrogen cooled

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HgCdTe photomixer, and a 64-channel spectral line receiver, located at the Coude focus of the 48" telescope at the Goddard Space Flight Center optical site (lat. = 39° 0.3'N, long. = 76° 52.6'W). Two sets of electronic filter banks provided high resolution coverage (5 MHz or 0.000167 cm^{-1})and low resolution coverage (50 MHz or 0.00167 cm^{-1}) with a total bandwidth of 1 GHz (0.03 cm^{-1}). The field of view of a heterodyne receiver is diffraction limited ($A\Omega \sim \lambda^2$) and was $\sim$ 4 arc-sec. The use of a CO_2 laser as a local oscillator limits atmospheric measurements to those lines which lie within the electrical bandwidth ($\pm$ 1.5 GHz) of the photomixer. A more detailed description of the instrument, of its advantages, and of its limitations has been given elsewhere (Abbas, et al. 1976, 1977; Mumma, et al. 1977).

In heterodyne detection, the infrared radiation from the source at frequency ν is mixed with radiation from the local oscillator (LO) at frequency ν_o, and the difference frequency signal in a bandwidth B both below and above the LO frequency is detected. This is referred to as double-sideband detection (DSB) in this paper. The DSB signal at frequency $\delta\nu = |\nu - \nu_o|$ corresponds to

$$I_{\nu}^{DSB}(\delta\nu) = I_{\nu}(\nu_o + \delta\nu) + I_{\nu}(\nu_o - \delta\nu) \qquad 1)$$

where I_{ν} is the source infrared radiance. The intensity in a single sideband may be retrieved by subtracting the corresponding mirror-sideband contribution. This may be done trivially if the mirror-sideband has no absorption lines and the continuum has the same level in both

sidebands. Inversion of observed spectral lines may then be carried out in the usual way (Abbas, et al. 1977).

On the other hand, if the mirror-sideband contains some additional lines of the absorbing gas, sideband-stripping requires detailed modelling of the monochromatic synthetic spectrum. It is sometimes more convenient to analyze the double-sideband spectrum directly. For solar observations, a double-sideband transmittance (τ^{DSB}) may be defined by dividing Eq. (1) by 2 I_o, where I_o is the continuum intensity above the atmosphere.

$$\tau_\nu^{DSB} = \frac{1}{2} [\tau_\nu (\nu_o + \delta\nu) + \tau_\nu (\nu - \delta\nu)] \qquad 2)$$

A synthetic plot of the earth's monochromatic absorption spectrum for a model atmosphere (Selby and McClatchey, 1975) in the frequency range 1043.113 cm^{-1} to 1043.213 cm^{-1} is shown in Fig. 1. Atmospheric CO_2 and O_3 absorption lines are identified on the plot. The moderately strong line (arrow) which lines within 1 GHz of the P(24) CO_2 line is the ozone line with upper and lower state quantum numbers (J',K_a',K_b'; J'',K_a'',K_b'') identified as (7,1,6; 7,1,7) with line strength S = 0.026 cm^{-1} $(cm\text{-}atm)^{-1}$.

The stratospheric ozone measurements reported here were made in the solar absorption mode on April 7, 1977 near noon (zenith angle $\sim 34^o$) with the CO_2 laser tuned to the P(24) line in the 9.4μm band (ν_o = 1043.1632 cm^{-1}). The integration time was ~12 minutes. Plots of the observed double-sideband spectrum measured with high and low resolution filter banks are shown in Fig. 2. The RF mixing frequency was chosen such that the observed O_3 line was

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centered on the high resolution filter bank. The low resolution plot shows the same O_3 line and its mirror image, along with the atmospheric CO_2 line.

The atmospheric O_3 line is found to be at 430 ± 10 MHz from the CO_2 line (with $\nu_o = 1043.1632$ cm^{-1}), providing an accurate line center position for the O_3 line as $\nu_o = 1043.1775 \pm .00033$ cm^{-1}. This compares with the values $\nu_o = 1043.1880$ cm^{-1} (AFCRL molecular line parameter atlas, McClatchey et al., 1973), $\nu_o = 1043.1761$ cm^{-1} (Barbe et al., 1977) and $\nu = 1043.1830$ cm^{-1} (Menzies, 1976).

Since the atmospheric transmittance was not directly calibrated, an absolute transmittance scale was determined by normalizing to the value of τ^{DSB} at $\nu = 1043.16833$ cm^{-1} (250 MHz away from the CO_2 line center) computed for a mid-latitude winter model atmosphere (Selby and McClatchey, 1975). Total vertical water vapor content assumed in the model was 623 cm-atm (precipitable water = 0.50 cm), which corresponds to the observed radiosonde data reported by the National Weather Service for the time of observations.

The observed spectrum of Fig. 2 bears little resemblance to the synthetic spectrum of Fig. 1 for the model atmosphere, because the observed spectrum contains contributions from the mirror-sideband. The synthetic spectrum shows that the mirror sideband contains weak O_3 absorption lines and the continuum levels of the two sidebands are unequal. In view of the discussion following Eq. (1), it was more convenient to analyze the observed double-sideband spectrum directly.

The observed O_3 line (Fig. 2) was inverted using the radiative transfer methods discussed elsewhere (Abbas, et al. 1977). Suitable weighting functions based on this line alone cover a range of altitudes from $\sim$ 10-30 km (Fig. 7 of Ref. 4). The retrieved O_3 mixing ratio profile is shown in Fig. 3. The retrieved profile has been extrapolated and is shown as a dashed line below 10 km and above 30 km. The nominal (mid-latitude winter model, Selby and McClatchey, 1972) mixing ratio profile of O_3 is also shown for comparison. The observed and synthetic double-sideband spectra are shown in Fig. 4, indicating good agreement between the computed and the observed values.

The retrieved ozone profile corresponds to a total vertical O_3 content of 0.34 cm-atm, which compares with the nominal value of 0.398 cm-atm for the mid-latitude winter model atmosphere. The retrieved profile (altitude range 10-30 km) indicates a maximum for the volume mixing ratio at $\sim$ 24 km as against 30-35 km for the model profile. Since the maximum vertical ozone content is believed to occur between the winter and spring months (Bauer, 1975), the measured value is consistent with this trend.

The measurements reported here are of a preliminary nature. More comprehensive measurements, involving a number of absorption lines of ozone, and a study of its diurnal variation are planned in the near future. Such completely resolved stratospheric lines can be inverted to provide accurate concentration profiles of stratospheric gases and trace constituents. The general application of such measurements to other molecules is presently limited by lack of availability of

continuously tunable local oscillators, and the accuracy is limited by the quality of molecular line parameters. These limitations are being steadily removed, and infrared heterodyne measurements are expected to play an important role in stratospheric research.

ACKNOWLEDGEMENT

Assistance of Mr. Gary Burgess in carrying out the numerical computations and contributions of Mr's J. Faris, J. Guthrie, C. Lowe and Drs. J. Hillman and D. Jennings are gratefully acknowledged.

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List of Figures

Fig. 1 Synthetic absorption spectrum of the earth's atmosphere in the frequency range 1043.113 cm^{-1} to 1043.213 cm^{-1}. The arrow denotes the (7,1,6)-(7,1,7) O_3 line discussed in the text.

Fig. 2 Observed double-sideband spectra of O_3 and CO_2 lines in the solar absorption mode:

(a) High resolution plot of O_3 line (ν_o = 1043.1775) on the left,

(b) Low resolution plot of the O_3 line and the overlapped CO_2 P(24) line on the right. The frequency difference ($\Delta\nu$) is referred to the center of the observed O_3 line instead of the local oscillator, for convenience. Note that the double-sideband spectrum is symmetric about the LO frequency which results from a mixing of the IF signal with an RF oscillator and detection of the two sidebands.

Fig. 3 Retrieved O_3 profile obtained from an inversion of the observed absorption line of Fig. 1. The modelled mixing ratio profile is shown with a dashed line.

Fig. 4 Synthetic double-sideband spectrum computed from the retrieved O_3 profile of Fig. 3. The observed spectrum is also shown. The frequency scale displayed on the top is the IF frequency ($\delta\nu$). The transmittance at each $\delta\nu$ contains contributions from both sidebands of the monochromatic spectrum (i.e. $\nu_o + \delta\nu$ and $\nu_o - \delta\nu$ (see text). The scale displayed at the bottom indicates the frequencies of the monochromatic spectrum which contribute to the flux measured at $\delta\nu$.

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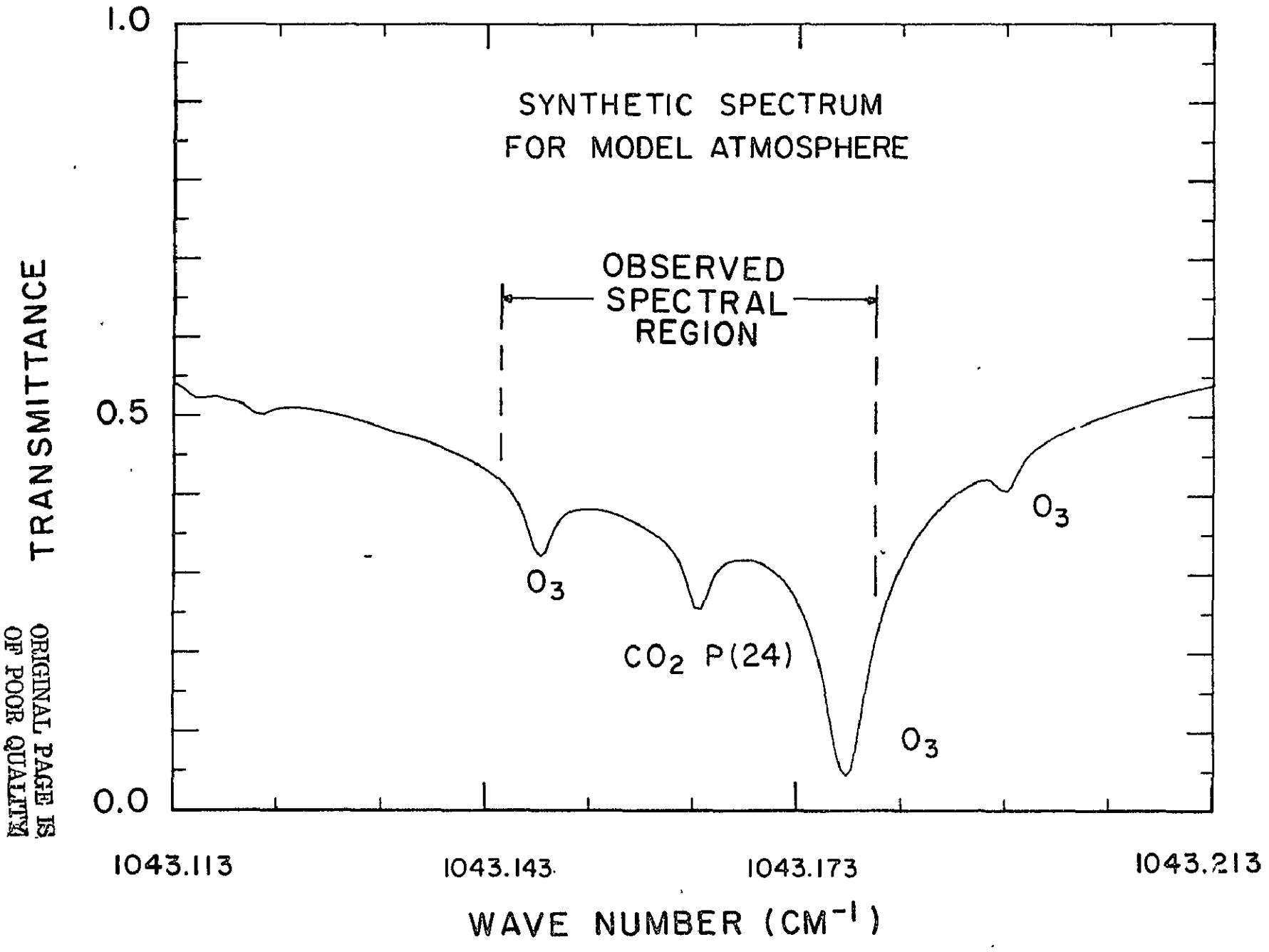

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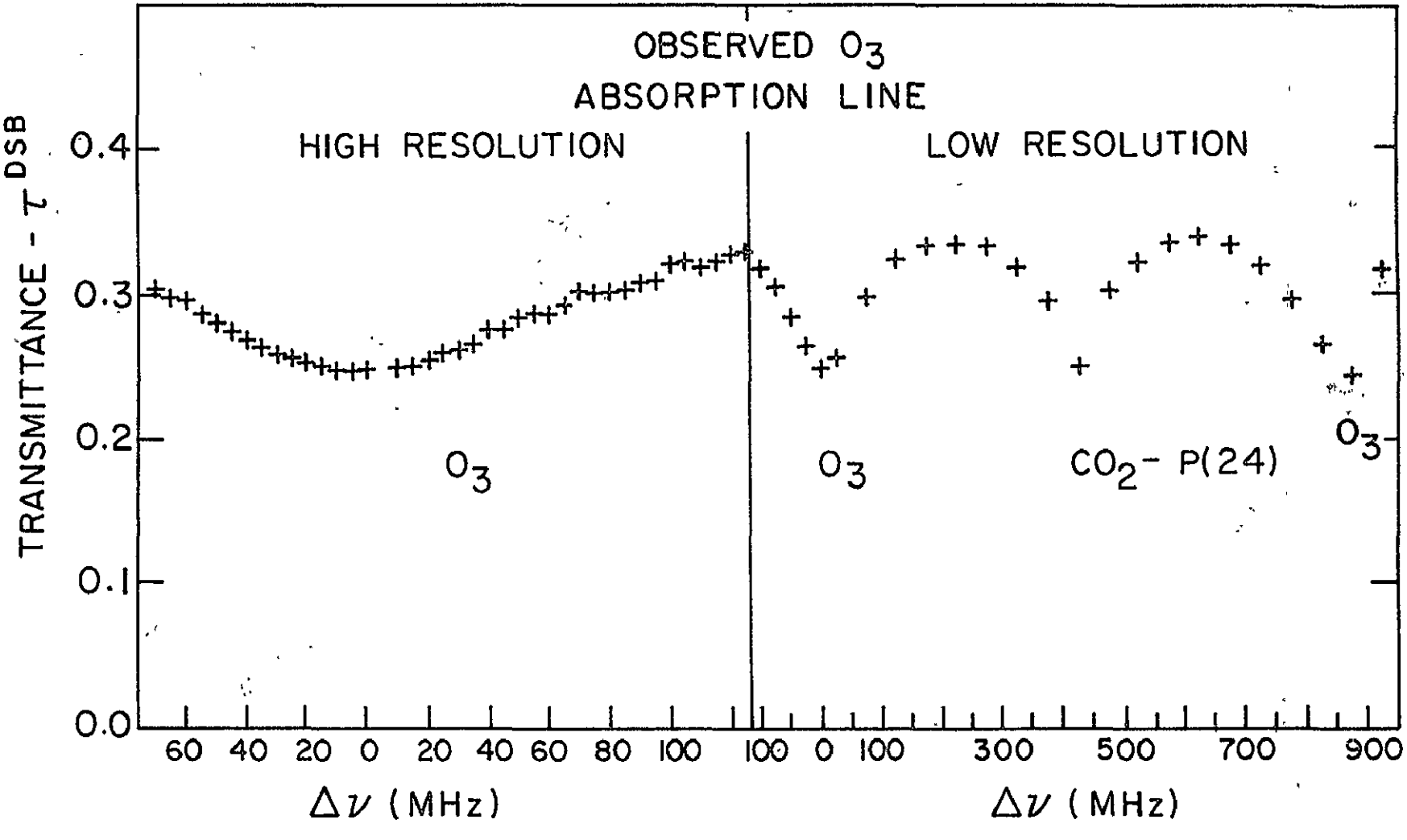
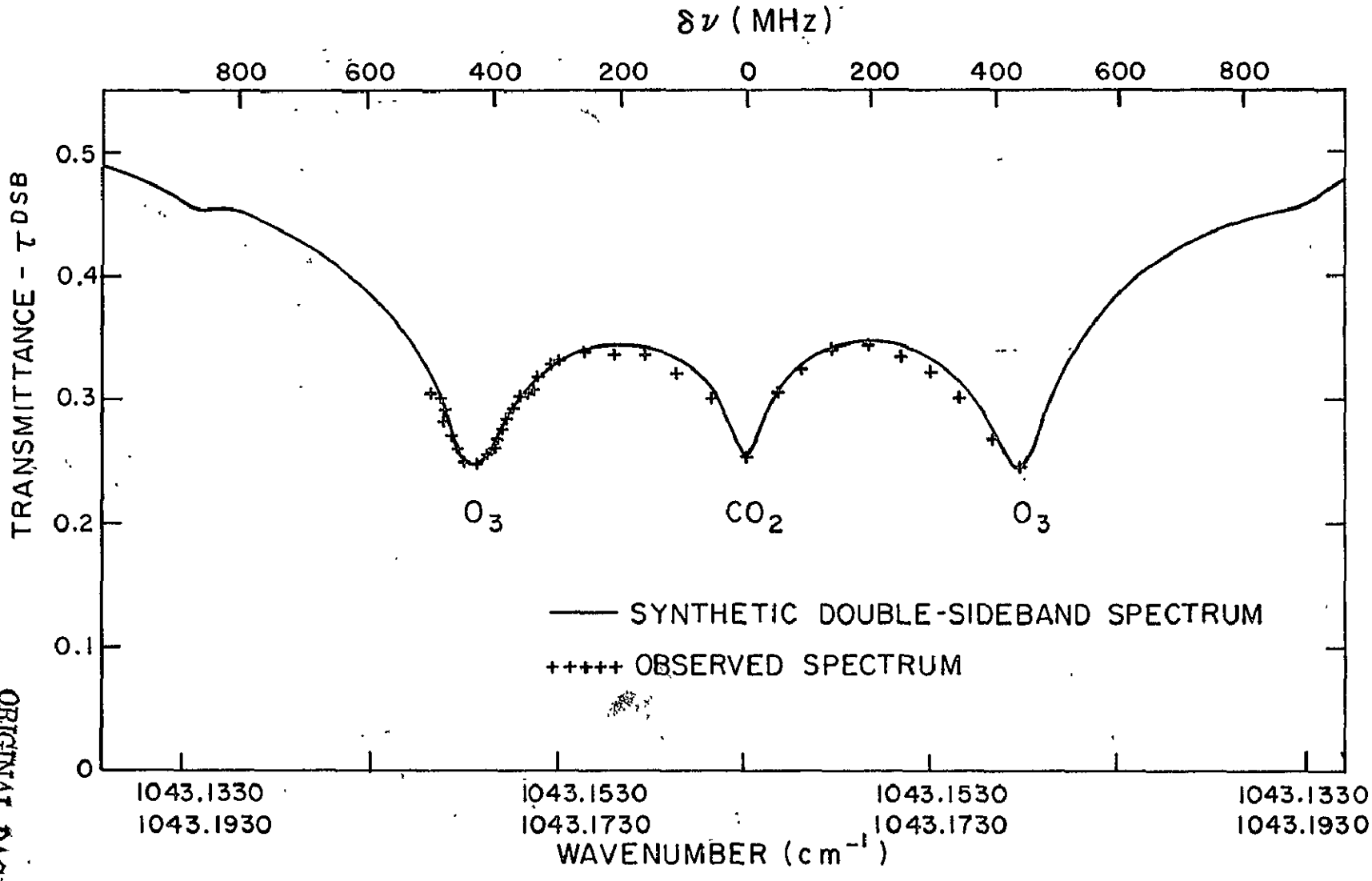

OBSERVED O_3
ABSORPTION LINE
HIGH RESOLUTION
LOW RESOLUTION
TRANSMITTANCE - τ^{DSB}
0.4
0.3
0.2
0.1
0.0
O_3
O_3
CO_2 - P(24)
O_3
60 40 20 0 20 40 60 80 100
100 0 100 300 500 700 900
$\Delta\nu$ (MHz)
$\Delta\nu$ (MHz)



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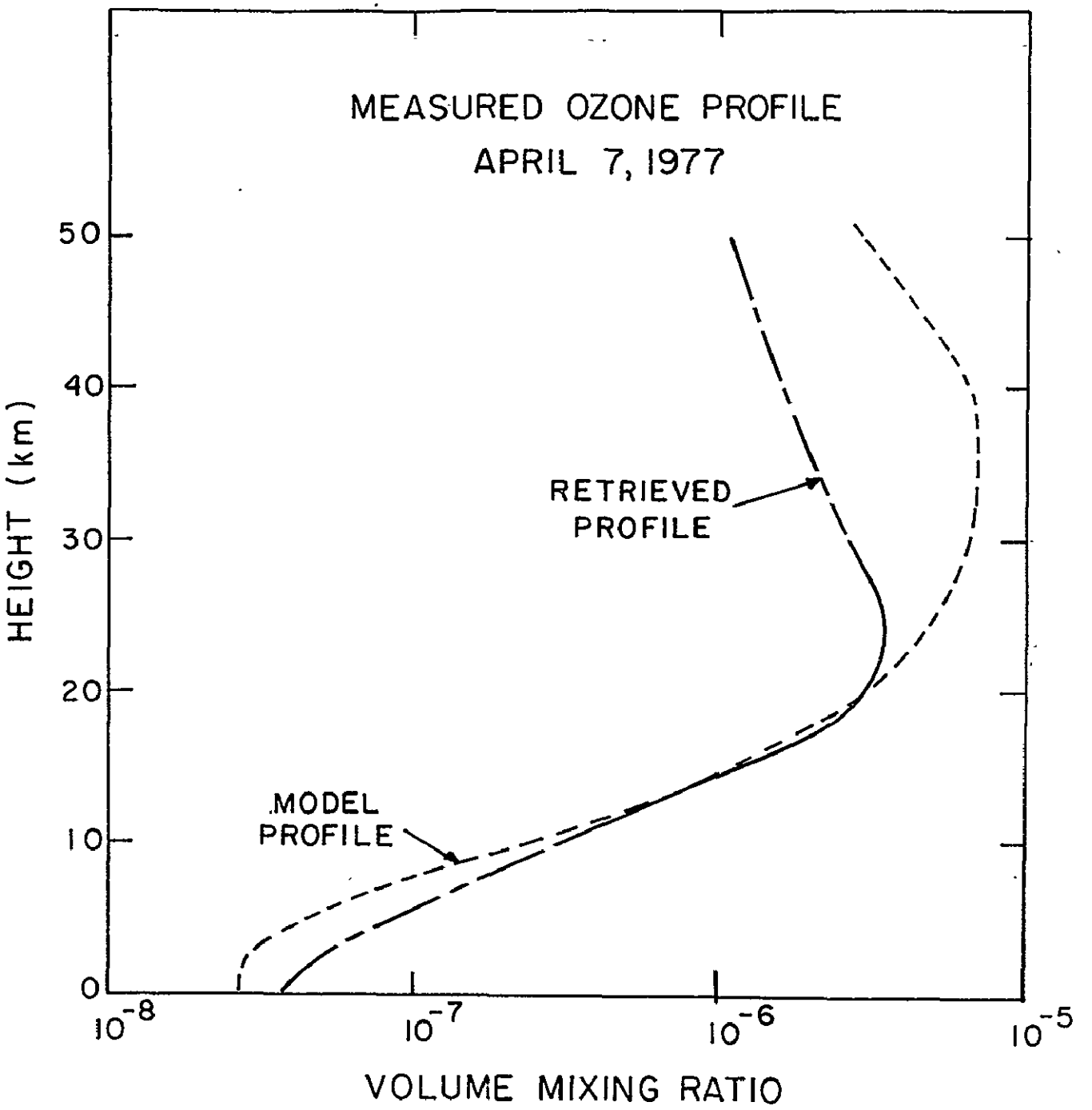
MEASURED OZONE PROFILE
APRIL 7, 1977
HEIGHT (km)
50
40
30
20
10
0
RETRIEVED PROFILE
MODEL PROFILE
10^{-8}
10^{-7}
10^{-6}
10^{-5}
VOLUME MIXING RATIO

BIBLIOGRAPHIC DATA SHEET

1. Report No.	2. Government Accession No.	3. Recipient's Catalog No.
TM 78067		

4. Title and Subtitle	5. Report Date
Stratospheric Ozone Measurement with an Infrared Heterodyne Spectrometer	6. Performing Organization Code

7. Author(s)	8. Performing Organization Report No.
Mian M. Abbas, Theodor Kostiuk, Michael J. Mumma, David Buhl, Virgil G. Kunde & Larry W. Brown	

9. Performing Organization Name and Address	10. Work Unit No.
NASA/Goddard Space Flight Center Greenbelt, MD 20771	11. Contract or Grant No.
	13. Type of Report and Period Covered
12. Sponsoring Agency Name and Address	14. Sponsoring Agency Code

15. Supplementary Notes

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A stratospheric ozone absorption line in the 10μm band has been measured and resolved completely, using an infrared heterodyne spectrometer with a spectral resolution of 5 MHz (0.000167 cm^{-1}). The vertical concentration profile of stratospheric ozone is obtained through an analytical inversion of the measured spectral line profile. The absolute total column density was 0.34 cm-atm with a peak mixing ratio occurring at ~ 24 km. The (7,1,6) - (7,1,7) O_3 line center frequency was found to be 1043.1775 ± 0.00033 cm^{-1}, or 430 ± 10 MHz higher than the P(24) CO_2 laser line frequency.

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17. Key Words (Selected by Author(s))	18. Distribution Statement

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price*

*For sale by the National Technical Information Service, Springfield, Virginia 22151.