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Stratospheric Ozone Measurement With An Infrared Heterodyne Spectrometer

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Greenbelt, Maryland 20771

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Mian M. Abbas*, Theodor Kostiuk, Michael J. Mumma,
David Buhl, Virgil G. Kunde, and Larry W. Brown

Laboratory for Extraterrestrial Physics
Infrared and Radio Astronomy Branch
NASA/Goddard Space Flight Center
Greenbelt, MD 20771

*Affiliated with Dept of Physics and Astronomy,
University of Maryland

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 ~ 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.

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 9.4μm 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

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 ~ 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 ~ 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.

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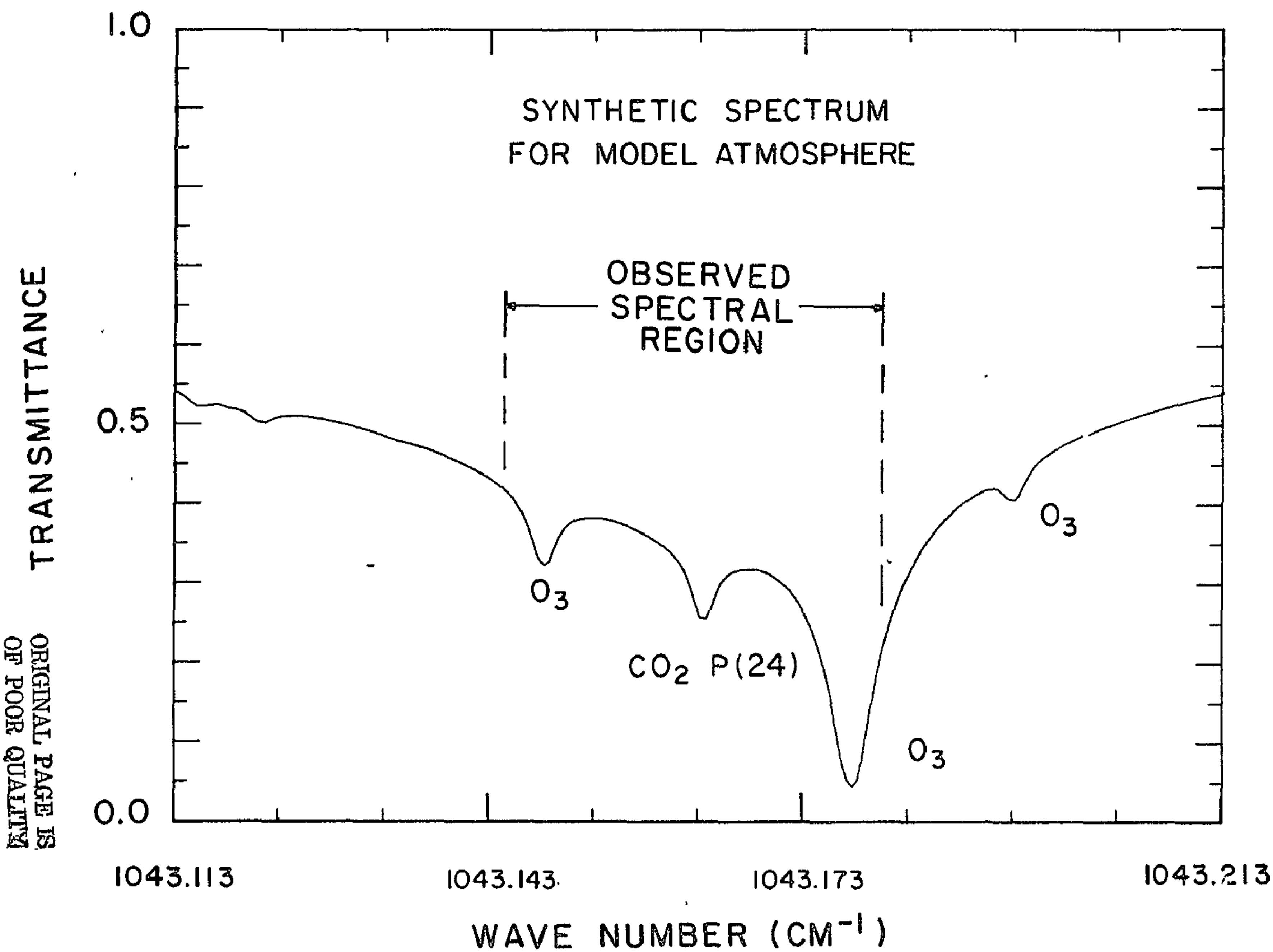
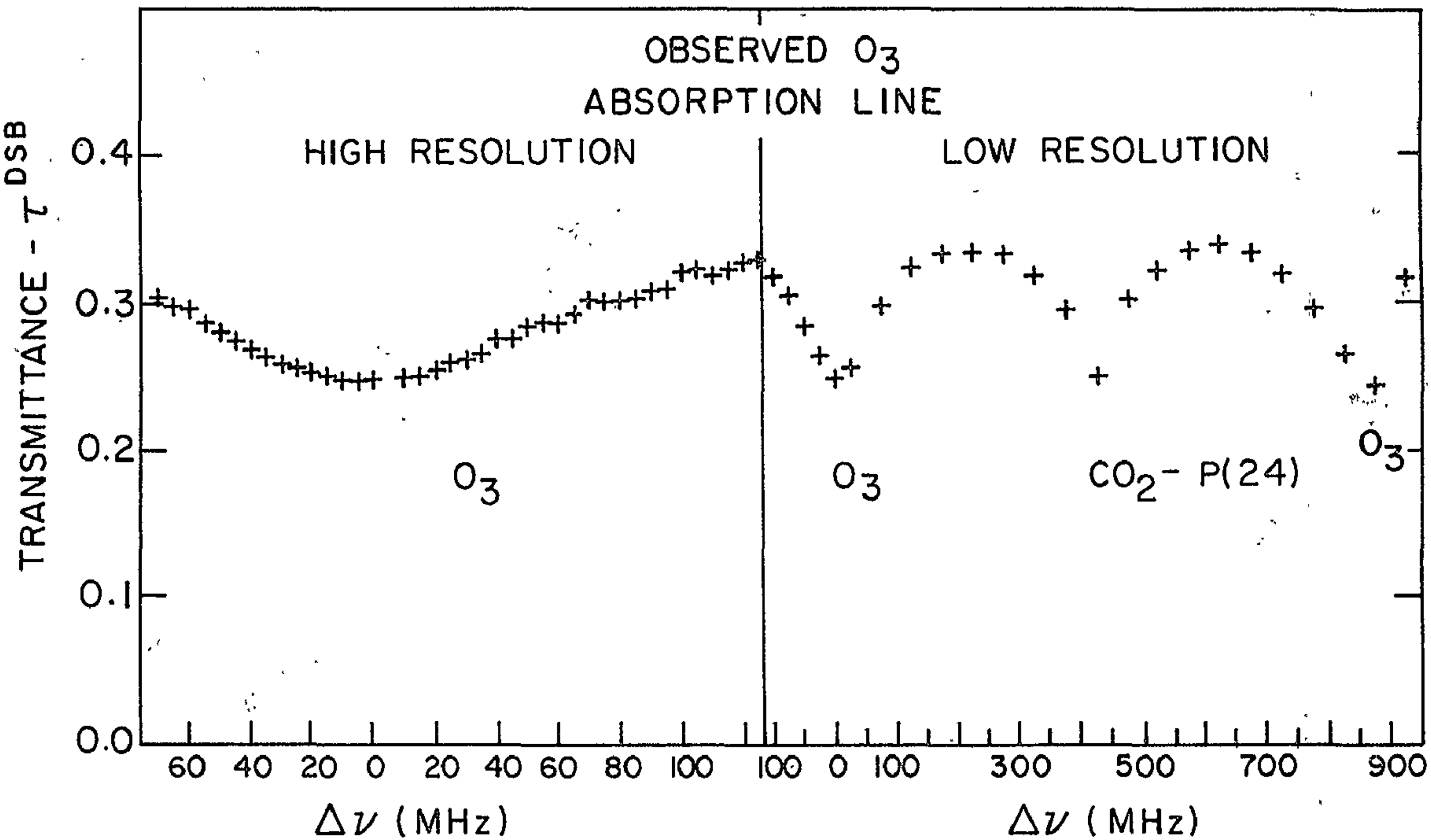
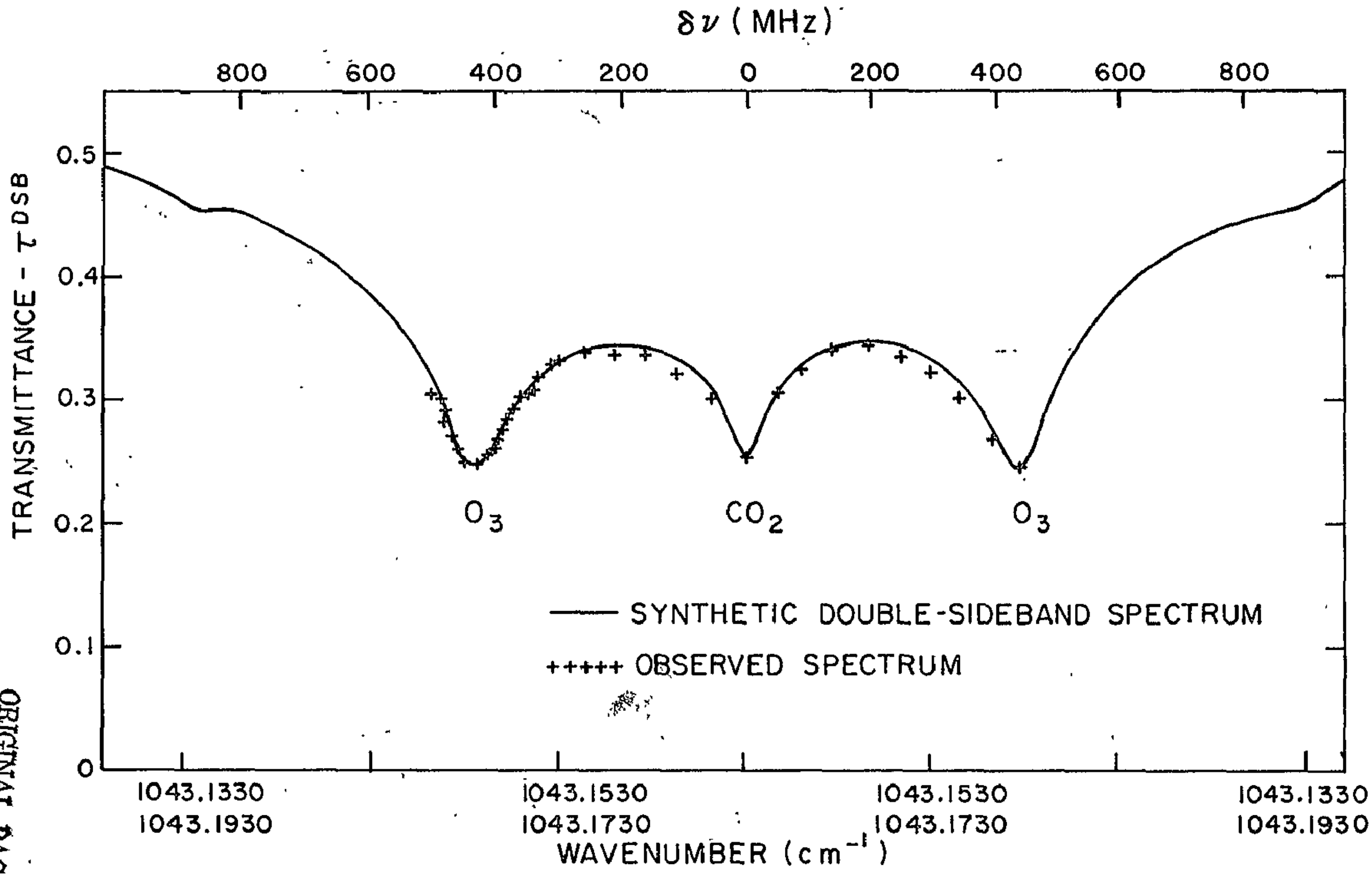
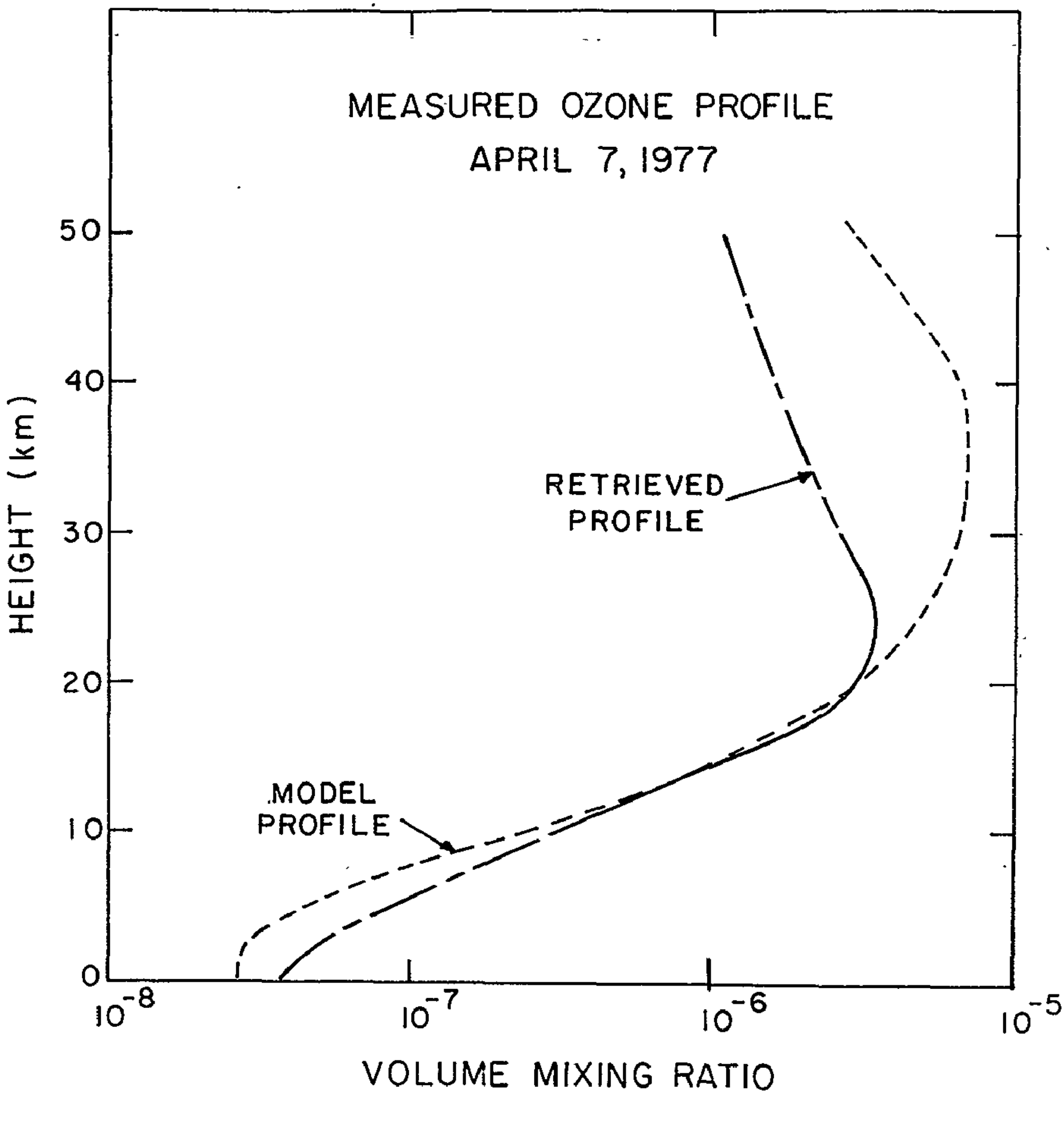

1.0
0.5
0.0
TRANSMITTANCE
SYNTHETIC SPECTRUM
FOR MODEL ATMOSPHERE
OBSERVED
SPECTRAL
REGION
O_3
CO_2 P(24)
O_3
O_3
1043.113
1043.143
1043.173
1043.213
WAVE NUMBER (CM^{-1})



OBSERVED O3
ABSORPTION LINE
HIGH RESOLUTION
LOW RESOLUTION
TRANSMITTANCE - τ DSB
0.4
0.3
0.2
0.1
0.0
O3
O3
CO2- P(24)
O3
60 40 20 0 20 40 60 80 100
100 0 100 300 500 700 900
Δν (MHz)
Δν (MHz)


δν (MHz)
800
600
400
200
0
200
400
600
800
TRANSMITTANCE - τ^DSB
0.5
0.4
0.3
0.2
0.1
0
O3
CO2
O3
SYNTHETIC DOUBLE-SIDEBAND SPECTRUM
+++++ OBSERVED SPECTRUM
1043.1330
1043.1930
1043.1530
1043.1730
1043.1530
1043.1730
1043.1330
1043.1930
WAVENUMBER (cm^-1)



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

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16. Abstract

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