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$^{13}CH_3OH$ IN OMC-1

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ABSTRACT

We report the identification of several previously unidentified or misidentified lines toward OMC-1 as components of the $J = 5 \rightarrow 4$ a-type band of $^{13}CH_3OH$, deduced by combining accurate laboratory spectroscopy with sensitive broad-band astronomical line searches. An LTE fit to the data yields a rotational temperature of $T_{rot} \sim 120$ K and a $^{13}CH_3OH$ column density of $\sim 1.3 \times 10^{15}$ cm^{-2}, averaged over a 30″ beam. A $^{12}C/^{13}C$ ratio of ~ 30 is derived. The assignment of the feature at 236063 MHz to the blended $K = \pm 2$ E transitions of $^{13}CH_3OH$, rather than to CO^+, removes a major discrepancy between observation and the predictions of ion-molecule chemical models of dense interstellar clouds.

Subject headings: interstellar: abundances — interstellar: molecules — line identifications

I. INTRODUCTION

Initially detected at 834 MHz toward the galactic center by Ball *et al.* (1970), methanol has since been observed in many galactic sources through more than fifty transitions in both its ground and first torsionally excited state (Gottlieb *et al.* 1979; Lovas *et al.* 1982; Johansson *et al.* 1984). As one of the most abundant interstellar molecules, methanol should be observable in various isotopic forms. Indeed, both $^{13}CH_3OH$ and CH_3OD have been detected in Sgr B2 (Gottlieb *et al.* 1979), but until very recently nowhere else. Johansson *et al.* (1984) have reported the detection of a single $^{13}CH_3OH$ line ($5_{-1} \rightarrow 4_0$ E) toward OMC-1 and deduce a $^{12}CH_3OH/^{13}CH_3OH$ ratio of ~ 30 from this transition.

A problem with hindered internal rotors such as CH_3OH is the difficulty of accurate prediction of the line frequencies. Laboratory spectroscopy is necessary to provide the accuracy required for interstellar molecule assignments, and in the case of $^{13}CH_3OH$ this information has been lacking in the millimeter-wavelength region. As a result of the interstellar and laboratory spectroscopy described here, it has become clear that $^{13}CH_3OH$ is a major contributor to the overall interstellar spectrum and that certain problems in the application of theoretical ion-molecule chemistry can be alleviated.

We report the detection of several components of the $J = 5 \rightarrow 4$, $\Delta K = 0$ band of $^{13}CH_3OH$ in OMC-1. Spanning some 50 cm^{-1} in upper-state energy, these lines permit a more reliable determination of the $^{12}C/^{13}C$ ratio than was possible with the single detection of Johansson *et al.* (1984). In addition, it is shown that the identifications of CO^+ and CH_3CHO in Orion by Erikson *et al.* (1981) are in fact most likely due to emission from $^{13}CH_3OH$ alone. This new interpretation is more consistent with current physical and chemical models of interstellar clouds.

II. OBSERVATIONS

The data were taken as part of a general spectral-line survey of OMC-1 in the 1.3 mm region using the No. 1 10.4 m telescope of the Owens Valley Radio Observatory (Sutton *et al.* 1985). The front end used was the 230 GHz superconducting tunnel junction (SIS) receiver constructed by Sutton (1983), while a 512 channel acousto-optic spectrometer (AOS) similar to that described by Masson (1982) served as the back end. The single-sideband receiver temperature was ~ 500 K at the $^{13}CH_3OH$ frequency of 236 GHz, and the AOS provided 527 MHz of coverage with 1.03 MHz resolution centered at an intermediate frequency of 1390 MHz.

Maps of the Moon were used to derive an efficiency of 0.85 for a spatially extended source, and an estimated beamwidth of 30″ at 230 GHz was determined from maps of planets. Antenna temperatures have been corrected for atmospheric and telescope losses using standard "chopper wheel" methods, and also by the extended source efficiency of 85%.

Raw spectra were obtained in a double-sideband mode by position-switching the telescope ±30′ in azimuth. The double-sideband survey data have been converted to a single-sideband spectrum by utilizing an algorithm similar in principle to the CLEAN algorithm (Högbom 1974) used to process interferometer maps (Sutton *et al.* 1985). This algorithm iteratively searches for the strongest features in the spectrum and removes their contributions, channel by channel, in the original double-sideband data. Spectra containing the $^{13}CH_3OH$ transitions were taken on 1982 November 8, 1983 January 2 and 6, and 1983 May 10 and 14. Spectra carefully chosen to be free of strong emission in the sideband opposite to $^{13}CH_3OH$ were also recorded as a check of the cleaning procedure. No notable differences were found between such double-sideband data and the cleaned single-sideband spectrum.

III. RESULTS AND DISCUSSION

a) $^{13}CH_3OH$

The spectrum of methanol is that of a nearly prolate asymmetric rotor, with the additional complication of hindered internal rotation. The intermediate torsional barrier height induces significant perturbations in the rotational spectrum, making accurate predictions of transition frequencies rather difficult. As a result, many transitions of methanol detected in interstellar clouds were, and still are, initially *unidentified*, even though several laboratory studies of methanol have been per-

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formed (Lees *et al.* 1973; Pickett *et al.* 1981; Sastry, Lees, and De Lucia 1984). This problem is especially acute in the near-millimeter-wave region, where highly excited lines may be quite strong (Sutton *et al.* 1985), and the lack of laboratory data is becoming a severe problem.

In this work $^{13}CH_3OH$ transition frequencies were initially calculated from the lower frequency data of Lees *et al.* (1973) and Haque *et al.* (1974) by using the internal rotor program of Herbst (1984). The errors in these predictions (0.4–0.6 MHz) were small enough to allow initial identification of the $^{13}CH_3OH$ transitions, but large enough to preclude definitive identifications and accurate estimates of the emission velocity. Rest frequencies of these transitions were therefore obtained with the Duke University millimeter-wave spectrometer, described in detail by De Lucia (1976).

Figure 1 presents a cleaned single-sideband spectrum of the region containing the $^{13}CH_3OH$ $J = 5 \rightarrow 4$ transitions. Not contained in this window are the two $K = 1$ A^- and A^+ components, which are split by about ± 2 GHz from the rest of the K-stack. Emission from at least three molecules, $^{13}CH_3OH$, HCO_2CH_3, and $^{34}SO_2$, is plainly visible in the figure. Table 1 summarizes the observations presented in Figure 1 along with some useful laboratory parameters. Rest frequencies for the two $^{34}SO_2$ lines are taken from Lovas (1983), while the methyl formate data are from an extensive laboratory study recently completed by Plummer *et al.* (1984). Averaging fits of unconstrained Gaussian components to all the isotopic methanol transitions gives $\langle v_{LSR} \rangle = 7.9 \pm 0.7$ km s^{-1} and $\langle \Delta v \rangle = 3.9 \pm 0.6$ km s^{-1}. These values are in excellent agreement with the large body of methanol measurements previously made in Orion (Hollis *et al.* 1983 and references therein). Line widths for the $^{13}CH_3OH$ fits listed in Table 1 were therefore constrained to 3.9 km s^{-1}, but were left unconstrained for the other species.

Excitation of the isotopic methanol components has been analyzed with an LTE model previously utilized by Linke, Frerking, and Thaddeus (1979). For optically thin emission from an unresolved source of angular diameter ϑ_S studied with a beam size ϑ_B, we may write

$$\ln\left(\frac{\vartheta_B}{\vartheta_S}\right)^2 \frac{3kc}{8\pi^3} \frac{\int T_A^* dv}{v^2 \mu^2 S} = \ln \frac{N_{COLM}}{Q(T_{ROT})} - \frac{E_u}{kT_{ROT}} = \ln \frac{N_u}{g_u} \quad (1)$$

where N_{COLM} is the total column density of $^{13}CH_3OH$ molecules; $Q(T_{ROT})$ is the rotational partition function; N_u, g_u, and E_u are the upper-state column density, degeneracy, and energy; and T_{ROT} is the temperature that characterizes the assumed Boltzmannian population distribution. The frequency, dipole moment, and line strength of the transition are denoted by v, μ, and S. In the high-temperature limit Q may be replaced by $Q = 2Z$, where Z is the usual rigid asymmetric rotor partition function, since the A and E symmetry species have approximately equal populations in this limit. For cool clouds, however, methanol is better described by analyzing the A and E species separately, since the lower zero-point torsional energy of the A symmetry species can create differences in the two populations.

Figure 2 graphically presents the results of a fit of equation (1) to the data using an assumed source size of 30″. Recent mapping of the high-energy $7_2 \rightarrow 8_1$ A^- transition by Olofsson (1983) characterizes the Orion methanol source as a spatially confined cloudlet whose position and v_{LSR} correspond closely to the 25″ × 11″ 6 cm formaldehyde emission region mapped with the VLA by Johnston *et al.* (1983). Lower energy transitions are clearly more extended, however (Johansson *et al.* 1984). Hills, Pankonin, and Landecker (1975) estimate a methanol source size of 30″ from mapping of the 25 GHz masers, a value used subsequently by many authors to model emission from the CH_3OH transitions detected in Orion. Numerically, our data are best fitted by the constants $T_{ROT} = 120 \pm 20$ K (1 σ) and $N_{COLM} = 1.3 \times 10^{15}$ molecules cm^{-2}, although the data appear consistent with temperatures in the range 100–200 K.

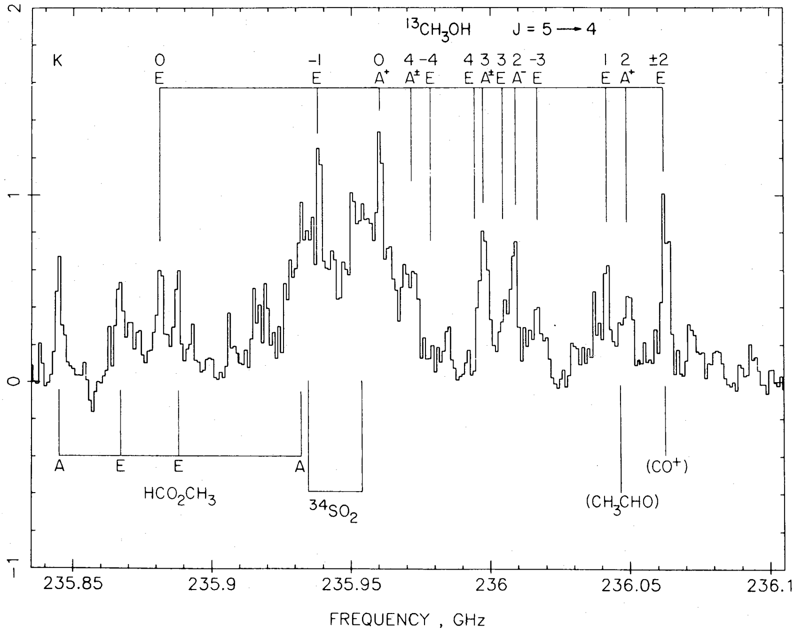

FIG. 1.—Spectrum of the $^{13}CH_3OH$ $J = 5 \rightarrow 4$ transitions observed toward OMC-1 with 1 MHz resolution. The frequency scale is derived by assuming a velocity with respect to the local standard of rest of 8 km s^{-1}. Lines associated with emission from CO^+ and CH_3CHO by Erikson *et al.* (1981) are indicated in parentheses.

TABLE 1

SUMMARY OF MOLECULAR EMISSION DETECTED IN THE $^{13}CH_3OH$ $J = 5 \rightarrow 4$, $\Delta K = 0$ BAND

Transition	Laboratory Frequency (MHz)	E_{upper} (cm^{-1})	S	T_A^* (K)	ΔV ($km\ s^{-1}$)
		$^{13}CH_3OH$			
$5_1 \rightarrow 4_1\ A^+$	234011.58(5)[a]	33.536[b]	4.80	0.76(12)[c]	3.9(6)
$5_0 \rightarrow 4_0\ E$	235881.17(5)	32.722	5.00	0.60(12)	3.9(6)
$5_{-1} \rightarrow 4_{-1}\ E$	235938.22(5)	27.509	4.80	0.68(12)	3.9(6)
$5_0 \rightarrow 4_0\ A^+$	235960.37(5)	23.615	5.00	0.71(12)	3.9(6)
$5_4 \rightarrow 4_4\ A^\pm$	235971.07(5)	79.770	3.60	0.25(12)	3.9(6)
$5_{-4} \rightarrow 4_{-4}\ E$	235978.62(5)	85.022	1.80	$\lesssim 0.12$	3.9(6)
$5_4 \rightarrow 4_4\ E$	235994.42(5)	90.637	1.80	0.72(12)	4.6(7)
$5_3 \rightarrow 4_3\ A^\pm$	235997.23(5)	58.403	6.40		
$5_3 \rightarrow 4_3\ E$	236006.10(5)	56.951	3.20	0.35(12)	3.9(6)
$5_2 \rightarrow 4_2\ A^-$	236008.39(5)	49.890	4.20	0.65(12)	3.9(6)
$5_{-3} \rightarrow 4_{-3}\ E$	236016.55(5)	67.364	3.20	0.36(12)	3.9(6)
$5_1 \rightarrow 4_1\ E$	236041.40(5)	38.255	4.80	0.56(12)	3.9(6)
$5_2 \rightarrow 4_2\ A^+$	236049.52(5)	49.893	4.20	0.41(12)	3.9(6)
$5_{-2} \rightarrow 4_{-2}\ E$	236062.00(5)	41.694	4.20	0.92(12)	3.9(6)
$5_2 \rightarrow 4_2\ E$	236062.85(5)	39.158	4.20		
$5_1 \rightarrow 4_1\ A^-$	237983.38(5)	33.933	4.80	0.84(12)	3.9(6)
		HCO_2CH_3			
$19_{7,13} \rightarrow 18_{7,12}\ A$	235844.50(5)[d]	100.806	21.90	0.54(12)	3.7(8)
$19_{7,13} \rightarrow 18_{7,12}\ E$	235865.87(10)	100.806	21.90	0.48(12)	4.1(8)
$19_{7,12} \rightarrow 18_{7,11}\ E$	235887.18(10)	100.811	21.90	0.54(12)	3.5(8)
$19_{7,12} \rightarrow 18_{7,11}\ A$	235932.34(5)	100.811	21.90	0.47(12)	3.2(8)
		$^{34}SO_2$			
$5_{2,4} \rightarrow 4_{1,3}$	235927.55(11)[e]	16.140	2.13	0.59(0.15)	24.9(1.5)
$10_{3,7} \rightarrow 10_{2,8}$	235951.97(11)	49.947	5.48	0.71(0.15)	26.7(1.5)
		CO^+			
$N = 2 \rightarrow 1, J = 3/2 \rightarrow 1/2$	235789.64(5)[f]	3.93	0.67	<0.25	...
$N = 2 \rightarrow 1, J = 5/2 \rightarrow 3/2$	236062.55(5)	3.94	1.20	...	...
2		CH_3CHO			
$12_{1,11} \rightarrow 11_{1,10} E$	235997.02(10)[g]	45.09	77.405	...	...
$12_{1,11} \rightarrow 11_{1,10} A$	236049.11(10)	45.04	77.405	...	...

NOTE.—Uncertainties in the values are given in parentheses and refer to the last digit(s) shown.

[a] This work.

[b] Upper-state energies for both the A and E symmetry species of $^{13}CH_3OH$ are referenced to the $J_K = 0_0$ A level.

[c] Blended with HCO_2CH_3 $16_{9,7} - 16_{8,8}$ A and $16_{9,8} - 16_{8,9}$ A transitions.

[d] Plummer *et al.* 1984.

[e] Lovas 1983.

[f] Sastry *et al.* 1981.

[g] Cohen and Blake 1984.

The fitted value of T_{ROT} is the same, within errors, as the value of 140 K derived for CH_3OH by Johansson *et al.* (1984) using a similar beam size, and is consistent with a gradual trend in excitation temperature seen in a number of other methanol millimeter and centimeter observations. Lovas *et al.* (1982) and Hollis *et al.* (1983), for example, find $T_{ROT} \sim 90$ K from an analysis of the low-lying $J = 2 \rightarrow 1$ ground-state lines, while the high-energy torsionally excited emission detected by these authors and by Sutton *et al.* (1985) indicates an excitation temperature nearer to 200 K. Transitions with moderate excitation requirements, such as those presented here, would be expected to be characterized by temperatures between these two extremes, as is observed. Errors in the derived column density are more difficult to estimate but would stem mainly from errors in the assumed source size.

The intensities of the parent and isotopic $J = 5 \rightarrow 4$, $\Delta K = 0$ bands give a direct $CH_3OH/^{13}CH_3OH$ ratio of ~ 15, which clearly suffers from saturation of the CH_3OH transitions (Sutton *et al.* 1985). Earlier estimates of the ^{12}C methanol column density using LTE techniques range from $3\text{–}7 \times 10^{16}$ cm^{-2}, with a value near 5×10^{16} being the most common (Hollis *et al.* 1983; Johansson *et al.* 1984). Thus the data are compatible with a $CH_3OH/^{13}CH_3OH$ ratio of 25–55 in Orion, with a most likely value of ~ 40.

If, as suspected, methanol is formed primarily through the radiative association reaction $CH_3^+ + H_2O$ followed by electron recombination (Gottlieb *et al.* 1979), then the $CH_3OH/^{13}CH_3OH$ ratio should be determined primarily by the $^{12}C^+/^{13}C^+$ ratio. Since differential photodissociation effects are likely to be unimportant in dense clouds, possible

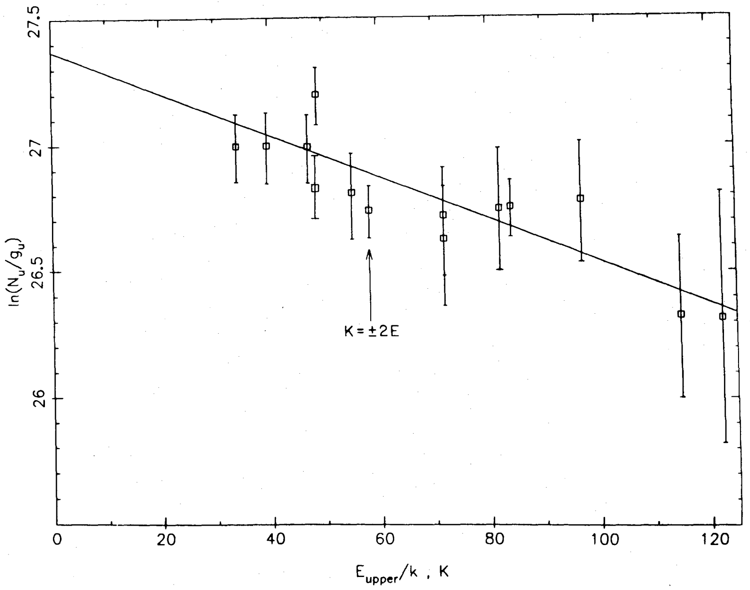

FIG. 2.—LTE rotation diagram of the observed $^{13}CH_3OH$ transitions, with data points centered in 1 σ error bars. The best fit, shown by the solid line, gives $T_{ROT} = 120$ K and $N_{COLM} = 1.3 \times 10^{15}$ cm^{-2}. The $K = \pm 2\ E$ transitions identified earlier as CO^+ are indicated by the arrow.

isotope fractionation may be estimated by using the analysis of Watson, Anicich, and Huntress (1976) from the relation

$$\left[\frac{^{12}C}{^{13}C}\right]_0 = e^{-35/T}\left[\frac{^{12}C}{^{13}C}\right]_{mol} , \qquad (2)$$

where T is the kinetic temperature (K), $[^{12}C/^{13}C]_0$ is the carbon ratio in all forms in the interstellar medium, and $[^{12}C/^{13}C]_{mol}$ is the observed molecular carbon ratio for molecules formed from C^+. This equation neglects the effects of depletion onto grains. For the observed methanol ratio, an overall gas-phase $^{12}C/^{13}C$ ratio of ~30 is indicated. Within errors, this agrees with the fairly low molecular $^{12}C/^{13}C$ values also derived in HCN, HC_3N, and OCS (Johansson *et al.* 1984) toward Orion, but is in sharp contrast to the higher $^{12}C/^{13}C$ ratios predicted for such molecules from large-scale time-dependent computer models (Langer *et al.* 1984).

b) CO^+

The fractional abundance of CO^+ in the interstellar medium was predicted to be quite low because of the reaction of CO^+ with the dominant constituents of both diffuse and dense molecular clouds, namely,

$$CO^+ + H \rightarrow H^+ + CO , \qquad (3)$$

$$CO^+ + H_2 \rightarrow HCO^+ + H . \qquad (4)$$

Both reactions are very rapid at 300 K (Karpas, Ancich, and Huntress 1979). The detection of CO^+ by Erickson *et al.* (1981) in Orion through emission from the $N = 2 \rightarrow 1$ transition therefore came as somewhat of a surprise, since the inferred fractional abundance, 10^{-10}–10^{-12}, was approximately 1–3 orders of magnitude larger than the predicted maximum fractional abundance of 10^{-13} (Herbst and Klemperer 1973; Prasad and Huntress 1980). Measurement of the reaction rate of CO^+ and H_2 between 100 and 390 K by Huntress *et al.* (1982) ruled out any temperature variation of reaction (4), leading these authors to conclude that CO^+ could not be present at the detected amounts in equilibrium with either H_2 or H. The narrow emission-line width rules out formation in shocked regions, and the reaction of C^+ with OH, O_2, or CO_2 in diffuse clouds produces insufficient amounts of CO^+ because of the small column densities and rapid photodissociation of the neutral precursors in C II regions (Erickson *et al.* 1981; Huntress *et al.* 1982). In short, it is difficult in theory to produce detectable amounts of CO^+ in any interstellar environment without invoking special conditions.

The data presented here, and the results of Erickson *et al.* (1981), are consistent with another interpretation of the 236063 MHz emission line. As shown in Figure 1 and Table 1, the average of the $K = \pm 2$ E-type transitions of $^{13}CH_3OH$ at 236062.43 MHz lies only 120 kHz from the CO^+ $N = 2 \rightarrow 1$, $J = 5/2 \rightarrow 3/2$ transition at 236062.55 MHz (Sastry *et al.* 1981). Figure 2 shows that the strength of this transition is in good agreement with the other methanol components; no excess emission appears to be present. Erickson *et al.* (1981) also find a weak feature which would be assigned to the $J = 3/2 \rightarrow 1/2$ component of CO^+ at 235789.64 MHz (Sastry *et al.* 1981). There may be a very weak bump at 235790.6 MHz in our cleaned spectrum ($T_A^* \sim 0.25$–0.30 K), but it is not evident in the proper sideband of any of the double-sideband data and could well be a cleaning residual. If this line were in fact due to CO^+, the relative line strengths of the $N = 2 \rightarrow 1$ components would imply that at least 0.45–0.60 K of the 236063 MHz feature must be due to CO^+. This would leave only ~0.3 K for the $K = \pm 2\ E$ $^{13}CH_3OH$ transitions, which have the highest expected strength of all the detected isotopic methanol lines.

The compelling chemical arguments listed above and the coincidence of transitions from a known, and abundant, interstellar molecule strongly suggest that the emission feature ascribed to CO^+ at 236063 MHz in Orion is in fact due primarily to $^{13}CH_3OH$. This interpretation is also consistent with the nondetection of the 236063 MHz feature in Sgr B2 by Erickson *et al.* (1981), since the lower cloud temperature favors population of the lowest K levels. Noise levels in the double-sideband data at the $N = 3/2 \rightarrow 1/2$ CO^+ component imply an upper limit for the CO^+ fractional abundance of

$f(CO^+/H_2) \lesssim 10^{-12}$, in better agreement with the standard ion-molecule synthetic models of interstellar clouds noted above. Other lines of CO^+ should be searched for to test this hypothesis and to set more stringent upper limits. The high atmospheric opacity at 118 GHz eliminates the $N = 1 \rightarrow 0$ transition, but the $N = 3 \rightarrow 2$ doublet near 354 GHz is available to ground-based systems.

c) CH_3CHO

Also illustrated in Figure 1 is the emission at 236045 MHz associated by Erickson *et al.* (1981) with the $12_{1,11} \rightarrow 11_{1,10}$ *A*-type transition of CH_3CHO. Table 1 shows that the rest frequency of this transition is actually 236049 MHz (Cohen and Blake 1984), which also corresponds to the $K = 2$ A^+ transition of $^{13}CH_3OH$. Similarly, the acetaldehyde *E*-type component at 235997 MHz is buried under the $^{13}CH_3OH$ $K = 3$ $A^{\pm}$ component. The strengths of these $^{13}CH_3OH$ components relative to the rest of the fit in Figure 2 implies, as for CO^+, that no other source of emission is present. We also failed to detect other $K_p = 1$ components of CH_3CHO in the spectral scan (Sutton *et al.* 1985), a finding consistent with the Onsala spectral survey of Orion (Johansson *et al.* 1984). We may, however, have detected weak emission from the $12_{0,12} \rightarrow 11_{0,11}$ transition near 226.5 GHz, from which an upper limit to the CH_3CHO column density of $\lesssim 5 \times 10^{13}$ cm^{-2} is derived with an assumed rotational temperature of 100 K. In any case, emission from CH_3CHO in Orion is extremely weak, if present at all.

Finally, it should be noted that the feature at 233213 MHz suggested as ethanol by Erickson *et al.* (1981) is in fact most likely due to emission from ethyl cyanide in the hot core. The line width and v_{LSR} are consistent with other hot-core species in Orion, since the rest frequency of the blended $26_{6,21} \rightarrow 25_{6,20}$, $26_{6,20} \rightarrow 25_{6,19}$, $26_{16,11} \rightarrow 25_{16,10}$, and $26_{16,10} \rightarrow 25_{16,9}$ transitions of ethyl cyanide is approximately 233207 MHz (Lovas 1983). In addition, no other transitions of ethanol have been detected in Orion by either the Onsala or the Owens Valley survey, whereas emission by C_2H_5CH is quite strong (Johansson *et al.* 1984; Sutton *et al.* 1985).

To summarize, several components of the $J = 5 \rightarrow 4$, $\Delta K = 0$ band of $^{13}CH_3OH$ have been detected toward OMC-1. An LTE analysis of the data implies an overall gas-phase $^{12}C/^{13}C$ ratio of ~ 30. Emission lines previously associated with CO^+ and CH_3CHO have been shown to be primarily or solely due to $^{13}CH_3OH$, improving the agreement between observation and theoretical models of dense interstellar clouds. Reinterpretations such as those presented here dramatically illustrate the immediate need for more complete laboratory information on even the most stable and basic interstellar molecules.

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