THE ASTROPHYSICAL JOURNAL, **270**:583–588, 1983 July 15

THE [HCO^+]/[HOC^+] ABUNDANCE RATIO IN MOLECULAR CLOUDS

R. C. WOODS AND C. S. GUDEMAN
Department of Chemistry, University of Wisconsin, Madison
R. L. DICKMAN, P. F. GOLDSMITH, G. R. HUGUENIN, AND W. M. IRVINE
Five College Radio Astronomy Observatory, University of Massachusetts, Amherst
AND
Å. HJALMARSON, L.-Å. NYMAN, AND H. OLOFSSON
Onsala Space Observatory, Sweden
Received 1982 June 14; accepted 1983 January 5

ABSTRACT

Using the recently determined rest frequency of its first rotational transition, we have made a sensitive search for the $J = 1 \rightarrow 0$ transition of HOC^+ in 14 interstellar sources. A line having a frequency and shape consistent with optically thin emission due to this species was detected toward Sgr B2; we provisionally identify this spectral feature as being due to HOC^+. Comparison with the $1 \rightarrow 0$ transition of $HC^{18}O^+$ in this source yields an isomeric abundance ratio $[HCO^+]/[HOC^+] \sim 330$, if the detected line is in fact due to HOC^+. Independent of, and consistent with, this conclusion, comparisons with isotopically substituted HCO^+ in the other clouds searched show that HCO^+ is at least two orders of magnitude more abundant than HOC^+. This large ratio places interesting constraints on the fractional abundance of related species.

Subject headings: interstellar: abundances — interstellar: molecules

I. INTRODUCTION

The existence of positive ions (HCO^+, N_2H^+, HCS^+) and of both members of the isomeric pair HCN/HNC in interstellar clouds provides strong support for ion-molecule schemes of molecule formation (e.g., Watson 1980). The ion HOC^+ is isoelectronic with HCN, HNC, N_2H^+, and HCO^+; and the recent laboratory measurement of the rest frequency for its $J = 1 \rightarrow 0$ rotational transition (Gudeman and Woods 1982) provides an opportunity to study both an additional interstellar ion and a second pair of isomers. Determination of the abundance ratio [HCO^+]/[HOC^+] can place important constraints on the fractional abundance of related interstellar species. We have searched for HOC^+ in both warm and cold interstellar clouds and have possibly detected it in Sgr B2. A preliminary analysis of the search results is presented, with emphasis on implications for abundances of other species.

II. OBSERVATIONS

Observations were made with the 14 m telescope of Five College Radio Astronomy Observatory (FCRAO) and the 20 m telescope of Onsala Space Observatory (OSO) between 1981 December and 1982 April. FWHM beamwidths at 89 GHz are 60″ and 42″, respectively. Both facilities employ cooled Schottky barrier diode mixers (Raisanen *et al.* 1981; Kollberg 1980) which achieve receiver temperatures ≤ 100 K (DSB) and 350 K (SSB) at FCRAO and OSO, respectively. The antenna temperatures reported in Table 1 have been corrected for radome and atmospheric losses, and for main beam coupling efficiency ($\eta_B = 0.6$ and $\eta_B = 0.55$ at FCRAO and OSO, respectively). The spectral line which we provisionally attribute to the $J = 1 \rightarrow 0$ transition of HOC^+, and that due to the $4_{04} \rightarrow 3_{03}$ transition of HCOOH, were clearly detected toward Sgr B2 in two separate sessions at FCRAO. On the second occasion, a local oscillator offset amounting to ~ 60 km s^{-1} was employed in order to unequivocally determine the line rest frequency, since the FCRAO receiver was tuned for double-sideband (DSB) response on both occasions.

III. RESULTS

Table 1 lists the sources searched, 5 σ upper limits to the main beam brightness temperatures T_R (except for the detection in Sgr B2), and calculated isomeric abundance ratios $R_1 \equiv [HCO^+]/[HOC^+]$, discussed below.

HOC^+ is a linear (or quasi-linear; cf. Gudeman and Woods 1982) closed-shell molecule, so that no fine or hyperfine structure is present to assist in a decisive confirmation that the line in Sgr B2, shown in Figure 1, is due to this ion. Both the observed V_{LSR} (approximately 60 km s^{-1} based on the measured HOC^+ rest frequency) and line shape agree with those of certain other molecular transitions in Sgr B2 believed to be optically thin (e.g., $C^{18}O$, HNCO, and H_2CO, Scoville,

TABLE 1

OBSERVATIONS OF HOC^+, $J = 1 \rightarrow 0$

Source	α(1950)	δ(1950)	T_R (K)	$H^{13}CO^+$ ($HC^{18}O^+$)	$R_1 = \frac{HCO^+}{HOC^+}$ (limit)
AFGL 437	03h30m31s	58°19′15″	0.08[a,b]	...	...
TMC-2	04 29 43	24 16 55	0.15[b,c]	0.7[d]	280
TMC-1(0,0)	04 38 38.6	25 36 18	0.15[b,c]	1.0[e]	400
(−4,6) ...			0.15[b,c]	1.0[e]	400
(−6,8) ...			0.15[b,c]	0.7[d]	280
Orion A	05 32 47	−05 24 20	0.15[f,g]	0.8[h]	210
				(0.08)[i]	260
NGC 2024	05 39 12	−01 55 42	0.12[a,b]	...	
	05 39 13.5	−01 56 57	0.10[a,b]	0.45[i]	270
L134N(CS)	15 51 24	−02 43 00	0.15[b,c]	0.4[d]	160
L134N(NH_3)	15 51 32.7	−02 42 51	0.4[a,g]	0.6[e]	90
				(0.1)[d]	120
ρ Oph A	16 23 22	−24 16 00	0.2[b,c]	1.7[j]	510
ρ Oph B	16 24 09	−24 21 49	0.2[b,c]	0.5[i]	150
W49N	19 07 49.8	09 01 17	0.15[f,g]	0.8[h]	320
W51N	19 21 22.4	14 25 13	0.3[f,g]	0.65[h]	130
W51M	19 21 26.2	14 24 43	0.2[f,g]	1.35[h]	400
ON 2	20 19 51.8	37 17 01	0.4[f,g]	0.5[h]	75
DR 21(OH)	20 37 14	42 12 00	0.2[f,g]	1.5[h]	450
DR 21	20 37 14.2	42 08 53	0.2[f,g]	(0.05)[k]	120
S140	22 17 42	63 03 45	0.3[f,g]	1.0[h]	200
Sgr B2	17 44 11	−28 22 30	= 0.08[b,f] ±0.04	(0.12)[k]	= 375

NOTE.—T_R is 5 σ upper limit on main beam brightness temperature, except for detection in Sgr B2; indicated error is 3 σ; data for $H^{13}CO^+$ and $HC^{18}O^+$ from indicated references; R_1 is lower limit on $[HCO^+]/[HOC^+]$, except for assumed detection in Sgr B2. We adopt $[HCO^+]/[H^{13}CO^+] = 20$ (galactic center), 40 (Orion), and 60 (remainder of galactic disk); and $[HCO^+]/[HC^{18}O^+] = 250$ (galactic center) and 500 (remainder of galactic disk) (Stark 1981; Penzias 1980).

[a] 250 kHz resolution.

[b] FCRAO.

[c] 100 kHz resolution.

[d] Guélin *et al.* 1982, Bell 7 m, $\Delta\nu = 62$ kHz.

[e] Average of data from d and i.

[f] 1000 kHz resolution.

[g] OSO.

[h] Downes *et al.* 1982, OSO, $\Delta\nu = 1$ MHz.

[i] Unpublished OSO data, $\Delta\nu = 70$ kHz ($H^{13}CO^+$) or $\Delta\nu = 1$ MHz ($HC^{18}O^+$).

[j] Wootten *et al.* 1982, NRAO 11 m, $\Delta\nu = 100$ kHz.

[k] Goldsmith and Linke 1981, and Stark 1981, Bell 7 m, $\Delta\nu = 1$ MHz (Sgr B2) or 250 kHz.

Solomon, and Penzias 1975; OCS, Goldsmith and Linke 1981). However, recent detection of the optically thin species $HC^{18}O^+$ and $HC^{17}O^+$ in Sgr B2 at $V_{LSR} \sim 54$ km s^{-1} (Guélin and Thaddeus 1979; Guélin, Chernicaro, and Linke 1982) would then appear to suggest some velocity disparity between the two formyl isomers in this source; this disparity may, however, simply be a consequence of low signal-to-noise ratio of the present data (Fig. 1) or of a velocity gradient in the source coupled to the factor of 2 difference in beam size (cf. Churchwell and Hollis 1983). An additional complication which hinders confident attribution of the line in Figure 1 to HOC^+ is the fact that the $10_{1,9} \rightarrow 10_{1,10}$ transition of HCOOH occurs at 89,490 MHz. While this transition has a line strength only ~5% that of the $4_{04}-3_{03}$ line shown in Figure 1 and discussed below (Willemot *et al.* 1980), Dr. B. E. Turner has informed us that anomalously strong, high-frequency *b*-type formic acid transitions have been previously seen toward Sgr B2 (although no published measurements are extant). However, if the spectral feature shown in Figure 1 is due to formic acid, it must be formed at a V_{LSR} some 11 km s^{-1} higher than the $4_{04}-3_{03}$ transition. This appears to be relatively unlikely.

In view of these ambiguities, decisive confirmation of interstellar HOC^+ must await detection of the $J = 3 \rightarrow 2$ (or higher) rotational transition, since absorption by atmospheric water vapor renders the $J = 2 \rightarrow 1$ transi-

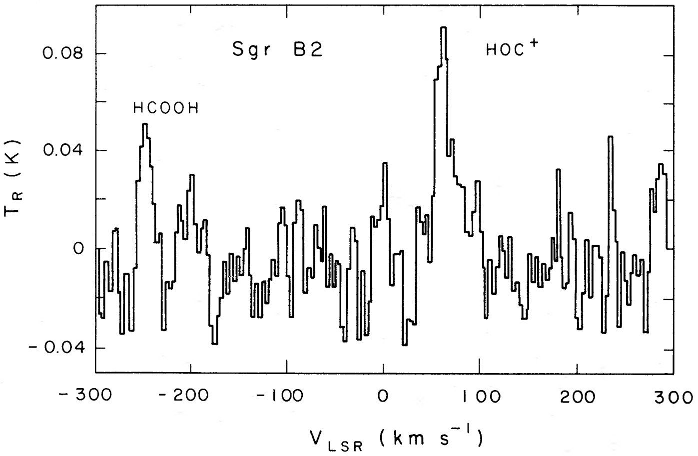

FIG. 1.—Spectrum toward Sgr B2 [$\alpha = 17^h44^m11^s$, $\delta = -28°22'30''$ (1950)] obtained at FCRAO with 1 MHz spectral resolution. Antenna temperature is corrected for antenna, radome, and atmospheric losses. Velocity scale assumes the measured rest frequency for HOC$^+$, $J = 1 \rightarrow 0$ ($\nu_0 = 89{,}487.414$ MHz), and we provisionally identify the labeled feature as the first interstellar detection of this ion (§ III). The $4_{04} \rightarrow 3_{03}$ transition of formic acid (HCOOH), with rest frequency 89,579.2 MHz, is also detected for the first time. Although the signal-to-noise ratio is clearly marginal, we note that the possible features at $V_{LSR} = -200$ km s^{-1} and 0 km s^{-1} correspond to a blend of CH_3CH_2CN $J = 10 \rightarrow 9$ transitions and to the CH_3OH $8_{-4} \rightarrow 9_{-3}$ E transition, respectively (Lovas *et al.* 1979). The emission at $V_{LSR} = 290$ km s^{-1}, corresponding to $\nu_0 = 89{,}419$ MHz, repeats in independent spectra and is presently unidentified.

tion unobservable from the ground. In what follows, we shall provisionally identify HOC$^+$ as the carrier of the spectral line shown in the center of Figure 1. However, it should be emphasized that the conclusions reached below concerning the extremely large isomeric ratio [HCO$^+$]/[HOC$^+$] would not be significantly altered if the detected line is indeed due to formic acid (or another species). If the 4 σ uncertainty in a single channel of the spectrum shown in Figure 1 is used in place of the observed peak line intensity, the isomeric ratio for Sgr B2 would increase to ~ 500.

The lower limits to the abundance ratio $R_1 \equiv$ [HCO$^+$]/[HOC$^+$] in Table 1 have been obtained by assuming that $J = 1 \rightarrow 0$ lines of HOC$^+$, H^{13}CO$^+$, and HC^{18}O$^+$ are optically thin and arise from the same spatial regions. Because of the large spontaneous emission rate for both species ($> 10^{-5}$ s^{-1} with μ[HOC$^+$] = 2.8 D, De Frees, Loew, and McLean 1982), excitation in the majority of the sources investigated will probably be subthermal. In this limit the column density $N \sim T_R$, not $\mu^2 T_R$ (cf. Linke *et al.* 1977). The calculated ratios R_1 listed in Table 1 clearly show that in all types of clouds investigated HOC$^+$ is at least two orders of magnitude less abundant than its isomer HCO$^+$.

In the case of Sgr B2, statistical equilibrium calculations were performed assuming equal collisional cross sections for HOC$^+$ and HC^{18}O$^+$ and a temperature and density derived from OCS observations (40 K and 10^4 cm^{-3}; Goldsmith and Linke 1981). The line intensities are well matched in the optically thin case for an abundance ratio $R_1 = 330$, close to the value calculated from a simple ratio of observed line intensities times isotopic abundances (Table 1), in agreement with the discussion of subthermal excitation in the preceding paragraph.

Figure 1 suggests the presence of other possible emission features, the most convincing of which (at $V_{LSR} \sim -250$ km s^{-1}) was also evident in the two independent local oscillator settings. We assign this second emission line to the $4_{04} \rightarrow 3_{03}$ transition of formic acid (HCOOH) with $\nu_0 = 89{,}579.2$ MHz. Formic acid has previously been reported in Sgr B2 (but not elsewhere in the interstellar medium) on the basis of centimeter-wavelength observations of the $1_{10} \rightarrow 1_{11}$ and $2_{11} \rightarrow 2_{12}$ transitions (Zuckerman, Ball, and Gottlieb 1971; Winnewisser and Churchwell 1975). The relative weakness of the $4_{04} \rightarrow 3_{03}$ transition confirms the suggestion by those authors that the low lying K-doublets are inverted. From the present data we calculate in the optically thin approximation $N(3_{03}) \sim 1.5 \times 10^{12}$ cm^{-2}; a rotational temperature of 40 K gives a total column density N(HCOOH) ~ 1×10^{14} cm^{-2}.

IV. DISCUSSION

The large values of the isomeric ratio R_1 found here provide important constraints on the abundances of related species. The chemistries of the three proton adducts HCO$^+$, HOC$^+$, and N_2H^+ are intimately related (Herbst *et al.* 1977; Snyder, Watson, and Hollis 1977); all three species are believed to be formed mainly by proton transfer from $H_3{}^+$. Destruction of HCO$^+$ is thought to be primarily via dissociative recombination with electrons, or by reactions with such neutral species as H_2O, OH, or Mg (collectively labeled here X) if these are sufficiently abundant (Herbst *et al.* 1976; Nobes and Radom 1981; Mitchell, Ginsburg, and Kuntz 1978; Prasad and Huntress 1980). We assume, based on a mix of experimental and theoretical evidence, that both HOC^+ and N_2H^+ will interact with CO via proton transfer to produce HCO^+, and $HOC^+ + N_2$ will similarly produce N_2H^+. Likewise, atomic hydrogen should react with HOC^+ to convert it to HCO^+, but not, because of symmetry, with the linear molecule N_2H^+. A model incorporating the above reactions is specified in Table 2. Quantities in square brackets are the densities of different species.

Treating only the relative abundances of HCO^+, HOC^+, and N_2H^+ as unknowns, the steady state rate balance from equations (2)–(16) in Table 2 has a fairly simple analytic solution. Let us make the reasonable assumptions that all recombination rates are equal ($k_4 = k_6 = k_7 \equiv k_r$) and that analogous ion-molecule reactions also have equal rates ($k_5 = k_8 = k_9$ and $k_{10} = k_{11}$). We obtain after some algebra

$$R_1 \equiv \frac{[HCO^+]}{[HOC^+]} = \frac{(k_{11}[CO] + k_{13}[H])}{(k_r[e] + k_5[X])}(1 + r_b + A\bar{r}_b) + \frac{k_{12}[N_2]}{k_r[e] + k_5[X]}(A + r_b + A\bar{r}_b) + (r_b + A\bar{r}_b) \geq 100 \qquad (1a)$$

and

$$R_2 \equiv \frac{[N_2H^+]}{[HOC^+]} = \frac{k_{12}(1 + \bar{r}_b)[N_2] + k_{13}\bar{r}_b[H]}{k_r[e] + k_5[X] + k_{10}[CO]} + \bar{r}_b \geq 10. \qquad (1b)$$

The numerical limit for R_1 is from Table 1, and that for R_2 is our estimate for molecular clouds from the $H^{13}CO^+$ and $HC^{18}O^+$ data in Table 1 and the N_2H^+ results given by Turner and Thaddeus (1977) and Snyder *et al.* (1977). The quantities A (a ratio of destruction rates), r_b (a generalized branching ratio for the formation of HCO^+ and HOC^+), and $\bar{r}_b$ (relating N_2H^+ and HOC^+ formation) are defined in Table 2.

Equations (1a) and (1b) provide important, new constraints on the relative abundances of species which are either not directly observable (N_2, e) or very difficult to observe (H_2O, H). Increasing concentrations of CO, N_2, and H I act to increase R_1, while larger electron and "water" abundances (cf. Table 2) favor a more equal isomeric ratio. In contrast, R_2 decreases as [CO] increases, emphasizing the coupled nature of the ion chemistry. The observational constraint that *both* R_1 and $R_2 \gg 1$ then may indicate that $[H] \gg [CO]$, in conflict with current theoretical estimates or assumptions (e.g., Mitchell, Ginsburg, and Kuntz 1978; Prasad and Huntress 1980; Graedel, Langer, and Frerking 1982).

TABLE 2

REACTIONS AND DEFINITIONS IN PRESENT MODEL[a]

Eq.	Reaction or Definition
2a	$CO + H_3{}^+ \rightarrow HCO^+ + H_2$
2b ...	$CO + H_3{}^+ \rightarrow HOC^+ + H_2$
3......	$N_2 + H_3{}^+ \rightarrow N_2H^+ + H_2$
4......	$HCO^+ + e \rightarrow CO + H$
5......	$HCO^+ + X \rightarrow$ products
6......	$HOC^+ + e \rightarrow CO + H$
7......	$N_2H^+ + e \rightarrow N_2 + H$
8......	$HOC^+ + X \rightarrow$ products
9......	$N_2H^+ + X \rightarrow$ products
10......	$N_2H^+ + CO \rightarrow HCO^+ + N_2$
11......	$HOC^+ + CO \rightarrow HCO^+ + CO$
12......	$HOC^+ + N_2 \rightarrow N_2H^+ + CO$
13......	$HOC^+ + H \rightarrow HCO^+ + H$
14a	$CO^+ + H_2 \rightarrow HCO^+ + H$
14b ...	$CO^+ + H_2 \rightarrow HOC^+ + H$
15a	$H_2O + C^+ \rightarrow HCO^+ + H$
15b ...	$H_2O + C^+ \rightarrow HOC^+ + H$
16......	$N_2^+ + H_2 \rightarrow N_2H^+ + H$
17......	$A \equiv k_{10}[CO]/(k_r[e] + k_9[X] + k_{10}[CO]) \leq 1$
18......	$r_b \equiv (k_{2a}[CO][H_3{}^+] + Y_a)/(k_{2b}[CO][H_3{}^+] + Y_b)$
19......	$\bar{r}_b \equiv (k_3[N_2][H_3{}^+] + Z)/(k_{2b}[CO][H_3{}^+] + Y_b)$
20......	$Y_i \equiv k_{14i}[CO^+][H_2] + k_{15i}[C^+][H_2O]\ (i = a, b)$
21......	$Z \equiv k_{16}[H_2][N_2{}^+]$

[a] X represents neutral species (the most abundant of which are likely to be H_2O, OH, and Mg) which may abstract a proton from HCO^+.

The following alternative explanations for the large values of R_1 and R_2 seem unlikely for the following reasons:

1. $r_b \gg 1$. It is probable that $k_{2a} \sim k_{2b} \sim k_3/2$, since the small dipole moment of CO will not be strongly oriented by the approaching $H_3{}^+$ ion. Then $r_b \sim 1$, a result supported by our laboratory experience concerning the relative strength of the HCO^+ and HOC^+ transitions, when we take account of the probable low-lying vibrational mode for the latter. However, recent experimental work (Illies, Jarrold, and Bowers 1982) indirectly suggests $k_{2b}/(k_{2b} + k_{2a}) = 6 \pm 5\%$, a value which would explain the present observations.

2. $[N_2] \gg [CO]$, which would make $\bar{r}_b \gg 1$. Both on the grounds of cosmic abundance and of detailed chemical modeling (e.g., Prasad and Huntress 1980), it seems likely that $\bar{r}_b \sim 2[N_2]/[CO] \leq 1$. An N_2 abundance somewhat greater than that for CO cannot be excluded observationally, however, and (particularly if r_b is greater than unity) this might in some cases explain the observed ratios R_1 and R_2.

3. $2[CO] \geq 100$ [H_2O, etc.] *and* $2[CO] \geq 3 \times 10^4[e]$. These are the constraints that follow from equations (1) in the simple model for which $(r_b, \bar{r}_b) \leq 1$, ($[N_2]$, [H]) < [CO], all relevant ion-molecule reactions proceed at similar rates k_L, and $k_r \sim 300\ k_L$—in other words, the model in which abundance differences between HCO^+ and HOC^+ are due to the faster destruction rate for HOC^+ caused by proton exchange to CO. *A priori* we had expected this model to be approximately correct. However, it is contradicted by the observation that $R_2 \equiv [N_2H^+]/[HOC^+] \gg 1$. Moreover, it seems quite unlikely that $2[CO] > 3 \times 10^4[e]$, except perhaps in cold dark clouds (Wootten, Loren, and Snell 1982; Dickman 1978; Guélin, Langer, and Wilson 1982); and $2[CO] > 100[H_2O]$ may not be satisfied in Orion (Waters *et al.* 1980; Phillips *et al.* 1978).

Although reliable observational estimates of atomic hydrogen abundance are notoriously difficult even for nearby dark clouds (cf. Mahoney 1972), values $[H]/[H_2] \sim 5 \times 10^{-3}$ are commonly deduced for such objects (Knapp 1974; McCutcheon, Shuter, and Booth 1978; Snell 1981). If indeed $[H] \gg [CO]$, this may place constraints on cloud ages, since the abundance of atomic hydrogen approaches its equilibrium value very slowly, according to present theoretical ideas (Allen and Robinson 1976).

It will be noted that in our model we have ignored the possibility of a reaction between H_2 and HOC^+ (radiative association). A reaction at the Langevin rate seems ruled out by recent experiments (W. T. Huntress, private communication) and is also unlikely on theoretical grounds (Nobes and Radom 1981). Additional quantitative data would be desirable. In addition, accurate determination of $[N_2H^+]$, particularly toward sources where HCO^+ and/or HOC^+ are known, would be very important to quantify the value of R_2 (or the analogous ratio to HCO^+). This will presumably require observation of a rare ^{15}N isotope of N_2H^+, to determine opacities and accurate column densities.

Finally, again assuming the identification of HOC^+ to be secure, we may estimate the column density and fractional abundance of this species in Sgr B2. Assuming an optically thin, homogenous slab with excitation temperature $T_{ex} \gg 2.7$ K, we find for the ground state $N_0(\text{Sgr B2}) \sim 4 \times 10^{11}\ \text{cm}^{-2}$, corresponding to a total column density $N(HOC^+) \sim 7 \times 10^{12}\ \text{cm}^{-2}$ if there is a uniform excitation temperature $T_{rot} = 40$ K. A statistical equilibrium calculation with $T_K = 40$ K and $n(H_2) = 10^4\ \text{cm}^{-3}$ gives essentially the same value. Goldsmith and Linke (1981) derive $[OCS]/[H_2] = 1 \times 10^{-9}$ and $N(HC^{18}O^+)/N(OCS) = 4 \times 10^{-3}$; together with the assumption (Table 1) that in Sgr B2 $[HCO^+]/[HC^{18}O^+] = 250$, we therefore find 3×10^{-12} for the fractional abundance of HOC^+ relative to H_2. In dark clouds such as TMC-1, assuming $5 \leq T_{ex} \leq 10$ K, $T_{rot} = T_{ex}$, and $N(H_2) = 10^{21}–10^{22}\ \text{cm}^{-2}$ (e.g., Elias 1978*a*, *b*), we obtain upper limits $N(HOC^+) \leq 2 \times 10^{11}\ \text{cm}^{-2}$ and $[HOC^+]/[H_2] \leq 10^{-10}$.

We are grateful to M. Guélin, G. Mitchell, W. Huntress, R. Nobes, and B. E. Turner for helpful comments. FCRAO is operated with support from NSF grant AST 81-21481 and by permission of the Metropolitan District Commission, Commonwealth of Massachusetts. OSO is operated with financial support from the Swedish Natural Science Research Council (NFR). Work at the University of Wisconsin was supported by the National Science Foundation Structural Chemistry Program. Visits by W. Irvine and R. C. Woods to OSO were supported by the Visiting Scientists grant from NFR and NSF grant INT-8205661. This is contribution number 516 of the Five College Astronomy Department.

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R. L. DICKMAN, P. F. GOLDSMITH, G. R. HUGUENIN, and W. M. IRVINE: Five College Radio Astronomy Observatory, GRC Tower B, University of Massachusetts, Amherst, MA 01003

C. S. GUDEMAN and R. C. WOODS: Department of Chemistry, University of Wisconsin, 1101 University Avenue, Madison, WI 53706

Å. HJALMARSON, L.-Å. NYMAN, and H. OLOFSSON: Onsala Space Observatory, S-43900 Onsala, Sweden