

Discovery of Interstellar Propylene (CH_2CHCH_3): Missing Links in Interstellar Gas–Phase Chemistry

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ABSTRACT

We report the discovery of propylene (also called propene, CH_2CHCH_3) with the IRAM 30-m radio telescope toward the dark cloud TMC-1. Propylene is the most saturated hydrocarbon ever detected in space through radio astronomical techniques. In spite of its weak dipole moment, 6 doublets (*A* and *E* species) plus another line from the *A* species have been observed with main beam temperatures above 20 mK. The derived total column density of propylene is $4 \times 10^{13} \text{ cm}^{-2}$, which corresponds to an abundance relative to H_2 of 4×10^{-9} , i.e., comparable to that of other well known and abundant hydrocarbons in this cloud, such as *c*- C_3H_2 . Although this isomer of C_3H_6 could play an important role in interstellar chemistry, it has been ignored by previous chemical models of dark clouds as there seems to be no obvious formation pathway in gas phase. The discovery of this species in a dark cloud indicates that a thorough analysis of the completeness of gas phase chemistry has to be done.

Subject headings: astrochemistry — line: identification — ISM: abundances — ISM: clouds — ISM: molecules

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1. Introduction

Cold ($T_{\text{kin}} \sim 10$ K), moderately dense ($n(\text{H}_2) \sim 10^4 \text{ cm}^{-3}$) dark clouds are the sites of low mass star formation. They exhibit a complex gas-phase ion-neutral chemistry leading to the formation of a large variety of molecules. The lack of internal heating sources and violent physical processes, like shocks, make these dense and quiescent cores the best sites to explore and to model interstellar gas-phase chemistry and molecular depletion into the dust grain surfaces. The dense filament TMC-1, embedded in the Heiles Cloud 2 (see, e.g., Cernicharo & Guélin 1987) in the Taurus region, is one of the richest molecular sources, with a large wealth of organic species such as unsaturated carbon chain radicals (C_nH) and cyanopolynes (HC_{2n+1}N). Whereas a common and well adopted view in the literature is to consider this region as dense, cold and quiescent (as indicated by the extremely narrow lines), some authors suggest that the presence of shocks could induce explosive injection of grain mantles and contribute to the formation of large carbon molecules such as methylcyano-diacetylene (Markwick et al. 2000; Snyder et al. 2006).

The history of molecular astronomy shows that interstellar molecules can be detected by specific searches, when frequencies are known and one has hints that a certain molecule is abundant. The most unexpected discoveries, however, resulted from spectral line surveys or were serendipitous. To our knowledge there are only two spectral line surveys of dark clouds, both in the direction of TMC-1. One was carried out using the Nobeyama radio telescope (Ohishi & Kaifu 1998; Kaifu et al. 2004) in the 8.8-50 GHz domain, with 414 detected lines arising from 38 different molecules, 11 of them unknown prior to the survey. Only two of the 38 species are diatomic (CS and SO), four are triatomic and the rest are polyatomic, the longest one having 11 atoms (HC_9N). The other survey was performed at lower frequencies, with the Arecibo radio telescope (Kalenskii et al. 2004), covering the range 4–6 GHz besides selected frequencies in the 8–10 GHz interval. They detected 29 lines arising from carbon-chain species such as C_4H , H_2C_4 , HC_3N , HC_5N , HC_7N , HC_9N , C_2S and C_3S .

Apart from CO and NO, all observed molecules in dark clouds usually have large dipole moments and most of them have rather low abundances (a few 10^{-10}). From this, one would naively expect that only polyatomic molecules with a large dipole moment should be detectable. Due to the low kinetic temperature of these clouds, heavy molecules will have their emission peak in the centimeter and millimeter wavelength domains. As a result of a 3 mm spectral line survey of several dark cloud cores (Marcelino et al. in prep.) and follow-up observations performed thereafter, we report the first detection of interstellar propylene (CH_2CHCH_3). This unexpected, nearly saturated hydrocarbon was detected toward TMC-1. This species, which is naively expected in hot cores rather than in dark clouds, has so far not

been considered in any chemical model of TMC-1 in spite of having an abundance similar to that of *c*-C₃H₂.

2. Observations

The observations were carried out with the IRAM 30-meter telescope¹ near Granada (Spain) Two SIS receivers sensitive in the 3 mm band were used simultaneously at the same frequency. Both receivers were tuned in single sideband mode with image rejections of ~ 20 dB. System temperatures were between 110–150 K during all observing periods. We used frequency switching mode with a frequency throw of 1.8 MHz (survey observations), while a throw of 7.2 MHz was used to obtain the new spectra outside the survey range in order to remove instrumental ripples and to improve baselines (see, Thum et al. 1995). The backend was the VESPA autocorrelator, with 40 kHz of spectral resolution (~ 0.13 km s⁻¹), providing a maximum effective bandwidth of 315 MHz. Intensity calibration was performed using two absorbers at different temperatures, and the atmospheric opacity was obtained from measurements of the sky emissivity and using the ATM code (Cernicharo 1985). On-source integration times were typically ~ 3 hours. Pointing and focus were checked on strong and nearby sources every 1.5 and 3 hours respectively.

The temperature scale is main-beam brightness temperature (T_{MB}). At the observed frequencies the beamwidth of the antenna is 24–29'' and the main beam efficiency is 0.78–0.75. The observed lines are shown in Fig. 1 and the line parameters are given in Table 1.

3. Results

During the 3 mm line survey of TMC-1, which was obtained in several observing runs between 2003 and 2007, we detected a pair of lines at 86.650 GHz with similar intensities and separated by 3 km s⁻¹, towards the cyanopolyne peak (CP) of this source [R.A.(J2000.0)=04^h 41^m 41.9^s, Decl.(J2000.0)=+25° 41' 27.1'']. These lines appear neither in the JPL nor in the CDMS catalogs (Pickett et al. 1998; Müller et al. 2001, 2005). Using the spectral line catalog developed and maintained at the DAMIR-IEM-CSIC (Cernicharo et al. 2000), we tentatively identified these two lines as one doublet arising from the (5₀₅ – 4₀₄) rotational transition (*A* and *E* species) of propylene (also called propene, CH₂CHCH₃) in the survey data of TMC-1. This molecule, whose structure is depicted in Fig. 2, is an internal rotor

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with a moderately high barrier against the torsion of the methyl group. This internal rotation produces a splitting of each rotational line by a value in the range of 0.3 MHz to 4 MHz, each member of the doublet belonging to the A and E species. Its rotational spectrum has been measured in the laboratory between 8–245 GHz (Wlodarczak et al. 1994) and up to 412 GHz (Pearson et al. 1994), which allows accurate frequency predictions within the frequency domain of our search. It has a small dipole moment, $\mu_a=0.36$ D, $\mu_b=0.05$ D (Lide & Mann 1957).

Encouraged by this tentative identification, we searched for other transitions of this molecule in the frequency range covered by our survey (85.9–93 GHz). The fact that the lines must appear as a doublet separated by 0.3–4 MHz, and having similar intensities helped us in the search. Using the mentioned catalog, which contains only complete predictions for the A species, together with the frequencies measured in the laboratory for the E species (Pearson et al. 1994), three more doublets at 87.1 GHz, 87.7 GHz and 90.0 GHz were identified at the 3–4 σ level. Director’s Discretionary Time at the 30-m telescope was used to confirm the identification of propylene in April and May 2007: We re-observed the latter three mentioned doublets and searched for three additional doublets lying outside the frequency range of the survey, at 84.1 GHz, 100.9 GHz and 103.7 GHz. We detected all the lines searched for, except for the E component of the $5_{15}-4_{14}$ transition at 84.1 GHz, for which the frequency is not known, since this transition was not measured in the laboratory by Pearson et al. (1994). However, since the separation in frequency of each doublet is small, the E line could be hidden by one hyperfine component of the $N = 1 - 0$ transition of ^{13}CCH at 84151.5 MHz (see top panel in Fig. 1). In fact, this hyperfine component of ^{13}CCH has a line strength twice less than that at 84153.4 MHz (also shown in the top panel of Fig. 1). Therefore, with a total of 13 observed lines, the first detection of interstellar propylene is definitively confirmed in TMC-1.

Line parameters were obtained from gaussian fits using the GILDAS package² and are given in Table 1. All the observed lines are centered at a systemic velocity around 5.6 km s^{-1} , which is in agreement with that derived from other molecules in TMC-1 (Kaifu et al. 2004). However, several lines show a weaker second peak at a higher velocity, that could arise from the emission of a different component of the cloud, at 5.9 or 6.1 km s^{-1} (Peng et al. 1998; Dickens et al. 2001). Line widths range from 0.3 to 0.6 km s^{-1} , being sometimes wider than the expected value of $\sim 0.4 \text{ km s}^{-1}$. The larger widths could imply again the emission from another component of the cloud.

Collisional rates are not available for this molecule. Hence we have used a standard

²<http://www.iram.fr/IRAMFR/GILDAS>

rotational diagram, including both species, to derive its column density and the rotational temperature, T_{rot} , of the energy levels. We have obtained $T_{\text{rot}} = 9.5 \pm 2.0$ K and $N(A\text{-CH}_2\text{CHCH}_3) = N(E\text{-CH}_2\text{CHCH}_3) = (2.0 \pm 0.8) \times 10^{13} \text{ cm}^{-2}$, i.e., a total column density for propylene of $(4.0 \pm 1.5) \times 10^{13} \text{ cm}^{-2}$. The rotational temperature is similar, within the errors, to that found for other species in TMC-1 (Ohishi & Kaifu 1998; Fossé et al. 2001; Kalenskii et al. 2004). However, the column density of propylene is particularly high for a saturated species and for a molecule having 9 atoms (compare Table 2). It is similar to that of $c\text{-C}_3\text{H}_2$ and to that of CH_3CCH , which also contain the methyl group and has a rather low dipole moment (see Table 2). It is worth to note that for some transitions the E species shows a slightly higher intensity than the A species (see Fig. 1 and Table 1), then the column density could be higher for the E species. In fact, performing separate rotational diagrams for the two species, the derived rotational temperature for the E species, $T_{\text{rot}} = 12$ K, is slightly higher than for the A , $T_{\text{rot}} = 8$ K. The total column density, however, do not differ much from the value obtained with a single rotational diagram. Hence, within experimental accuracy both species seem to have the same abundance.

4. Discussion

Propylene, a rather simple hydrocarbon, has escaped detection because apparently nobody ever considered a directed search worthwhile, and also because of its small dipole moment, 10 times smaller than that of other three carbon molecules (see Table 2). Pearson et al. (1994) already suggested that this molecule could be detected in space. However, as this hydrocarbon is nearly saturated, they pointed out that the best place to search for it could be hot cores in high-mass star forming regions. Prior to this detection, we had already searched for propylene in the spectral line survey of Orion (Tercero and Cernicharo, in preparation), but no lines were found within a detection limit of 50 mK. We have checked the U-lines in previous line surveys of Orion and Sgr B2 at different frequency bands (Sutton et al. 1985; Cummins et al. 1986; Turner 1989; Schilke et al. 1997; Nummelin et al. 1998; Friedel et al. 2004), and also the unidentified lines reported by Kaifu et al. (2004) towards TMC-1. None of these lines coincide with the predicted strongest lines of propylene.

The abundance of propylene relative to H_2 in TMC-1 is 4×10^{-9} , assuming that $N(\text{H}_2) = 10^{22} \text{ cm}^{-2}$ (Cernicharo & Guélin 1987). In spite of being a fairly abundant hydrocarbon, it has not been previously considered in chemical models of dark clouds. Reaction networks used to model interstellar chemistry are usually tested by comparing with the molecular abundances observed in TMC-1 (Smith et al. 2004; Woodall et al. 2007). Thus, much effort has been done to explore and include reactions leading to the formation of the observed

molecules, the abundances of which are in general satisfactorily explained, although the chemistry of not observed molecules is far from complete.

We have searched in the literature for reactions relevant to the chemistry of propylene and performed chemical models to investigate the most likely formation pathways. The synthesis of many hydrocarbons in dark clouds involve a sequence of ion-molecule reactions with the dissociative recombination (DR) of a molecular cation and an electron as the last step (Herbst & Leung 1989). Thus, the DR of the $C_3H_7^+$ ion, which is known to produce $C_3H_6 + H$ with a branching ratio of 0.42 (Ehlerding et al. 2003), could be a key step in the synthesis of propylene. If so, the problem of producing propylene translates into how to form the ion $C_3H_7^+$. Condensation reactions of an ionic and a neutral hydrocarbon such as $C_2H_4^+ + CH_4 \rightarrow C_3H_7^+ + H$ as well as other similar ones, are endothermic, too slow or tend to produce products other than $C_3H_7^+$ (Anicich 2003). Consideration of radiative association reactions of hydrocarbon ions with H_2 is an interesting possibility when other channels are endothermic. We have checked that such a possibility occurs for $C_2H_2^+$ and larger hydrocarbon ions with 2, 3, 4 and 5 carbon atoms. However, the corresponding reaction rates are not available in the literature and different reaction channels may occur. Introducing these reactions with realistic rate coefficient values and subsequent dissociative recombination allows to build propylene at a reasonable level.

Propylene could also be one of the products in the DR of ions larger than $C_3H_7^+$. Recent storage ring experiments have shown that substantial fragmentation, with break of C-C bonds, occurs in the DR of hydrocarbon ions (Viggiano et al. 2005). For example, chemical models predict an efficient synthesis of the ion $C_6H_7^+$ in dark clouds, the DR of which is assumed to give benzene and C_6H_2 (both predicted with an abundance of $\sim 10^{-9}$; McEwan et al. 1999), although it could also produce smaller neutral fragments such as propylene. The synthesis of propylene through neutral-neutral reactions such as $CH_3 + C_2H_4$ or $C_2H_3 + CH_4$ is not effective as these reactions are endothermic.

Lastly, highly saturated hydrocarbons form easily in grain surfaces by direct hydrogenation, although it is not clear how these mantle-species would be desorbed to the gas phase at the low temperatures of dark clouds. A possibility is to involve shocks and their role in the release of saturated adsorbed species from the surface of the grains (Markwick et al. 2000), or involve a local enhancement of the cosmic ionization rate which will have an important effect on the chemistry and evolutionary status of TMC-1 (Hartquist et al. 2001). However, the detected column density of methanol in TMC-1 does not support this scenario. Furthermore, if this mechanism is working in TMC-1, other hot-core like molecules (CH_3OCH_3 , CH_3COH , ...) should be detectable there. Recent work on non thermal desorption mechanisms from dust grains (Garrod et al. 2007) show that some molecules formed on

grain surfaces could go to the gas phase. The presence of hydrocarbons such as propene is an important information to improve the models as the hydrogenation proceeds on grain surfaces and the degree of surface hydrogenation increases with time. In any case, it seems that gas-phase chemistry is missing important reactions involving small hydrocarbons, which could make molecules like propylene to have large abundances. The detection of propylene suggest that other small hydrocarbons, like propane, could be present in dark clouds. A search for it is currently undergoing with the IRAM 30-m telescope.

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Table 1. Line parameters of the observed transitions from gaussian fits

Rest Frequency ^a (MHz)	Species	Transition	E_{up} (K)	$\mu^2 S$ (D ²)	$\int T_{MB} dv$ (K km s ⁻¹)	V_{LSR} (km s ⁻¹)	Δv (km s ⁻¹)	T_{MB} (K)
84151.670 (7)	<i>A</i>	5 ₁₅ – 4 ₁₄	13.9	0.610	0.014 (4)	5.72	0.46	0.028 (4)
86651.566 (7)	<i>A</i>	5 ₀₅ – 4 ₀₄	12.5	0.635	0.027 (4)	5.52	0.62	0.041 (4)
87134.576 (6)	<i>A</i>	5 ₂₄ – 4 ₂₃	19.8	0.534	0.005 (2)	5.68	0.32	0.016 (3)
87677.584 (6)	<i>A</i>	5 ₂₃ – 4 ₂₂	19.8	0.534	0.007 (2)	5.65	0.41	0.017 (3)
89998.180 (7)	<i>A</i>	5 ₁₄ – 4 ₁₃	14.8	0.610	0.012 (2)	5.53	0.31	0.036 (3)
100911.394 (8)	<i>A</i>	6 ₁₆ – 5 ₁₅	18.8	0.741	0.010 (2)	5.62	0.43	0.022 (4)
103689.979 (8)	<i>A</i>	6 ₀₆ – 5 ₀₅	17.5	0.761	0.020 (2)	5.71	0.55	0.033 (4)
86650.873 (24)	<i>E</i>	5 ₀₅ – 4 ₀₄	12.5	0.635	0.017 (2)	5.68	0.35	0.044 (4)
87137.941 (6)	<i>E</i>	5 ₂₄ – 4 ₂₃	19.8	0.531	0.011 (2)	5.76	0.42	0.024 (3)
87673.663 (29)	<i>E</i>	5 ₂₃ – 4 ₂₂	19.8	0.531	0.008 (2)	5.68	0.42	0.018 (2)
89997.232 (13)	<i>E</i>	5 ₁₄ – 4 ₁₃	14.8	0.610	0.014 (2)	5.63	0.46	0.029 (3)
100911.141 (5)	<i>E</i>	6 ₁₆ – 5 ₁₅	18.8	0.741	0.018 (2)	5.72	0.56	0.031 (4)
103689.117 (35)	<i>E</i>	6 ₀₆ – 5 ₀₅	17.5	0.761	0.017 (2)	5.79	0.41	0.040 (4)

Note. — Number in parentheses are 1σ uncertainties in units of the last digits.

^aFrequencies for the *A* species are from Cernicharo et al. (2000) catalog, and those for the *E* species are the ones measured by Pearson et al. (1994).

Table 2. Column densities of several hydrocarbons in TMC-1

Species	μ^a (D)	N (cm^{-2})	Species	μ^a (D)	N (cm^{-2})
C_2H ⁽¹⁾	0.77	7×10^{13}	$c\text{-C}_3\text{H}_2$ ⁽²⁾	3.43	6×10^{13}
$l\text{-C}_3\text{H}$ ⁽²⁾	3.55	8×10^{11}	H_2C_4 ⁽⁴⁾	4.10	8×10^{12}
$c\text{-C}_3\text{H}$ ⁽²⁾	2.40	1×10^{13}	H_2C_6 ⁽⁵⁾	6.20	5×10^{11}
C_4H ⁽³⁾	0.87	3×10^{14}	CH_3CCH ⁽¹⁾	0.78	8×10^{13}
C_5H ⁽⁴⁾	4.88	9×10^{12}	$\text{CH}_3\text{C}_4\text{H}$ ⁽⁴⁾	1.21	1×10^{13}
C_6H ⁽²⁾	5.54	8×10^{12}	$\text{CH}_3\text{C}_6\text{H}$ ⁽⁶⁾	1.50	3×10^{12}
$l\text{-C}_3\text{H}_2$ ⁽²⁾	4.10	2×10^{12}	CH_2CHCH_3 ⁽⁷⁾	0.36	4×10^{13}

^aDipole moments are from JPL and CDMS catalogs

References. — (1) Pratap et al. (1997); (2) Fossé et al. (2001); (3) Guélin et al. (1982); (4) Ohishi & Kaifu (1998); (5) Langer et al. (1997); (6) Remijan et al. (2006); (7) This work

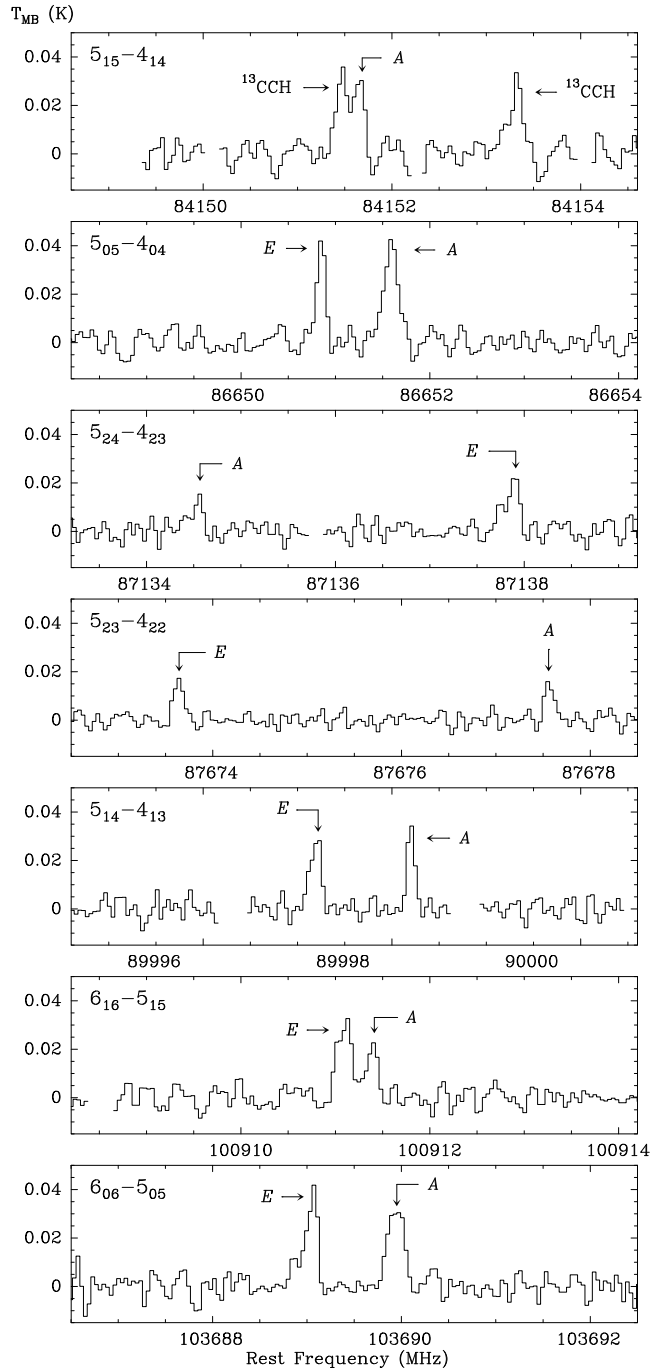


Fig. 1.— Observed transitions arising from both A and E species of CH_2CHCH_3 . Rest frequency is computed for an $v_{\text{LSR}} = 5.6 \text{ km s}^{-1}$. Note on the top panel that only the A species was detected (see text).

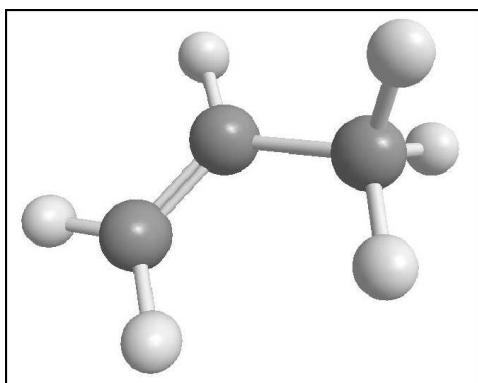


Fig. 2.— Molecular structure of Propylene. Spheres represent carbon (dark grey) and hydrogen (light grey) atoms.