

RADIO AND MILLIMETRE OBSERVATIONS OF LARGER MOLECULES

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ABSTRACT. The number of known interstellar molecules has increased steadily since 1970 and stands presently at 68. Of the molecules discovered in recent years more than half contain three or more heavy elements.

The number of sources where HC₅N or HC₃N has been detected now includes 21 dark clouds, four circumstellar shells, two bipolar nebulae, the line of sight to the Cas A supernova remnant, and possibly a comet. The more abundant large molecules are useful diagnostic probes, and preliminary statistical equilibrium calculations of the widespread new ring, C₃H₂, are presented which indicate its usefulness as an indicator of H₂ density.

1. INTRODUCTION

The search for new chemicals in the interstellar medium and circumstellar envelopes continues to be an important and productive pursuit of astronomers. From an astrophysical viewpoint, the two main goals of these searches are to determine molecular abundances and to provide diagnostic tools for the determination of physical conditions within molecular clouds.

In this review I shall first summarize developments in the search for new, larger molecules - i.e. those containing three or more heavy atoms. Then, in Section 3, some of the more recent astronomical observations are reviewed, and the importance of a number of these larger molecules as astrophysical probes is discussed.

2. NEW LARGE MOLECULES

2.1. Recent Discoveries

Table 1 contains a list of molecules with three or more heavy atoms which have been detected recently. Of these new, heavy molecules, all are carbon-rich and have been detected in TMC-1, except for SiCC which, to my knowledge, has not yet been sought there. Thus, TMC-1

continues to demonstrate a uniquely rich carbon chemistry. Experience has shown that any spectral line which is enhanced in TMC-1, compared to other sources, arises from a carbon-rich molecule.

TABLE 1. NEW LARGE MOLECULES RECENTLY DETECTED IN THE INTERSTELLAR MEDIUM

Mole- cule	Where Observed	Abundance Relative to H ₂	Ground State con- figuration	Molecular Constants (MHz)	Ref.
HC ₁₁ N	TMC-1 IRC10216	1.2x10 ⁻¹⁰ * 7x10 ⁻⁸	Linear ¹ Σ	B ₀ =169.2757 D ₀ =3x10 ⁻⁷ , μ=5.0D	1,2,3
CH ₃ C ₃ N	TMC-1	4.5x10 ⁻¹⁰ *	Symmetric Top	B ₀ =2065.7387 D _J =93.65x10 ⁻⁶ D _{JK} =19.925x10 ⁻³ μ=4.8D	2,4
CH ₃ C ₄ H	TMC-1 TMC-1 TMC-1	2x10 ⁻⁹ * 1.7x10 ⁻⁹ * 3.5x10 ⁻⁹	Symmetric Top	B ₀ =2035.746 D _J =6x10 ⁻⁵ D _{JK} =19.8x10 ⁻³ μ=1.21D	2,5, 6,7
C ₃ O	TMC-1	1.4x10 ⁻¹⁰	Linear ¹ Σ	B ₀ =4810.889 D _J =7.84x10 ⁻⁴ μ=2.39D	8,9
SiCC	IRC10216	1x10 ⁻¹⁰	Triangular Asymmetric Top	A=52390 B=13156.2 C=10447.4, μ=3.2D τ ₁ =-6.35, τ ₂ =-0.917 τ _{cccc} =-0.106	10
C ₃ H	TMC-1 IRC10216	5x10 ⁻¹⁰ 1.9x10 ⁻¹¹	Linear ² Π	A _{eff} =431093 B=11186.323 D=5.57x10 ⁻³ Y _{eff} =30.6 P=7.51, q=16.48 μ=3.1D	11
C ₃ H ₂	TMC-1 Sgr B2 Orion KL IRC10216	5x10 ⁻⁹ 9x10 ⁻¹¹ 5.4x10 ⁻¹¹ 4x10 ⁻¹²	Ring Asymmetric Top	A=35092.5964 B=32212.9312 C=16749.3147 μ=3.3D, τ ₁ =-0.6767 τ ₂ =-0.1628 τ _{aaaa} =-0.5901 τ _{bbbb} =-0.2982 τ _{cccc} =-0.0361	12,13
CH ₃ C ₅ N**	TMC-1	-	Symmetric Top	B ₀ =778.040	14

* Based on revised dipole moments from Snyder *et al.* (1985).

**Tentative detection - not yet confirmed

References: (1) Bell *et al.* (1982) (2) Snyder *et al.* (1985)

References - Cont'd: (3) Bell and Matthews (1985) (4) Broten et al. (1984) (5) Walmsley et al. (1984) (6) MacLeod et al. (1984) (7) Loren et al. (1984) (8) Brown et al. (1985) (9) Matthews et al. (1984) (10) Thaddeus et al. (1984) (11) Thaddeus et al. (1985a) (12) Thaddeus et al. (1985b) (13) Matthews and Irvine (1985) (14) Snyder et al. (1984)

As the list of carbon molecules found in TMC-1 grows, some patterns are emerging. Thaddeus et al. (1985a) have pointed out that acetylenic radicals, $\text{HC}\dots\text{C}$, with an even number of C atoms, tend to be significantly more abundant than those with an odd number. The reverse seems to be true for the cyanized carbon chains, HC_nN . In spite of the abundance of HCN and HC_3N , recent attempts to detect HC_2N have been unsuccessful (Feldman, 1985).

The observed column densities of the methylated polyynes decrease in TMC-1 by a factor of about three with the addition of two C atoms - $\text{N}(\text{CH}_3\text{C}_2\text{H})/\text{N}(\text{CH}_3\text{C}_4\text{H}) \approx 3$ and $\text{N}(\text{CH}_3\text{CN})/\text{N}(\text{CH}_3\text{C}_3\text{N}) \approx 3.5$ (Askne et al. 1984, Matthews and Sears, 1983, and Table 1). This is similar to the abundance decrease shown by successively larger cyanopolyynes, and MacLeod et al. (1984) have argued that this similar behaviour manifested by chain molecules with different terminal groups reflects the relative abundance of pure carbon chains. They have also estimated the abundances of the important but unobservable (at radio wavelengths) molecules, HC_2H and HC_4H . If the ratio $\text{N}(\text{HC}_3\text{N})/\text{N}(\text{CH}_3\text{C}_3\text{N}) \approx 65$ is taken as representative of the abundance difference between molecules terminated by H and by CH_3 , this implies that $\text{N}(\text{HC}_2\text{H}) \approx 65 \text{N}(\text{CH}_3\text{C}_2\text{H}) = 5.2 \times 10^{15} \text{ cm}^{-2}$ and $\text{N}(\text{HC}_4\text{H}) = 65 \text{N}(\text{CH}_3\text{C}_4\text{H}) = 1.1 \times 10^{15} \text{ cm}^{-2}$ in TMC-1. For $\text{N}(\text{H}_2) = 1 \times 10^{22} \text{ cm}^{-2}$ in TMC-1, the abundances relative to H_2 are $[\text{HC}_2\text{H}] = \text{N}(\text{HC}_2\text{H})/\text{N}(\text{H}_2) = 5 \times 10^{-7}$ and $[\text{HC}_4\text{H}] = 1 \times 10^{-7}$. A somewhat higher estimate of $[\text{HC}_4\text{H}]$ is obtained by assuming $\text{N}(\text{HC}_4\text{H})/\text{N}(\text{C}_4\text{H}) = \text{N}(\text{HC}_3\text{N})/\text{N}(\text{C}_3\text{N}) = 19$. For $\text{N}(\text{C}_4\text{H}) \approx 2.5 \times 10^{14} \text{ cm}^{-2}$, this gives $[\text{HC}_4\text{H}] = 5 \times 10^{-7}$, which further implies $[\text{HC}_2\text{H}] \approx 10^{-6}$ in TMC-1. (See references in MacLeod et al. 1984 for abundances).

Recent observations (Matthews and Irvine, 1985) suggest that the most important of the new molecules in Table 1 is C_3H_2 (cyclopropenylidene). Its structure is not linear, but triangular, and its spectrum that of an asymmetric top with b-type transitions (Thaddeus et al. 1985b). Matthews and Irvine (1985) have detected the $1_{01}-1_{10}$ line at 18343 MHz in most of the 25 varied objects where they looked for it. It is widespread throughout the Galaxy, and its large dipole moment makes it readily observable. These properties suggest that C_3H_2 will be an important probe of physical conditions in molecular clouds and we return to this topic in Section 3.

2.2. The Future

A survey of the literature shows that the number of known astrophysical molecules has increased linearly at an average rate of three per year since 1971. There are now at least 68 different compounds known, exclusive of isotopic variations, and, if the trend of the last 15 years continues, over 100 will be identified by the end of the century.

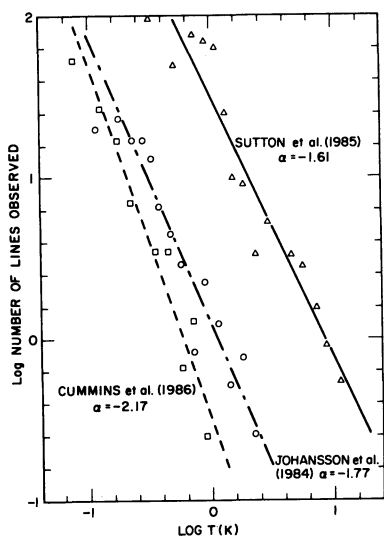


Fig. 1. The number of spectral lines, N , detected with antenna temperature in a range $T \pm \Delta T/2$ for three surveys.

Figure 1 illustrates how the number of observed lines increases with decreasing line strength in the three surveys. N is the number of lines observed with antenna temperature in an interval ΔT_A centred on T_A . For the two lower frequency surveys $\Delta T_A = 0.05\text{K}$; for the Sutton *et al.* (1985) survey, $\Delta T_A = 0.20\text{K}$. The less sensitive spectra in the lower frequency surveys were not used in preparing Figure 1 in order to obtain reasonably uniform sensitivity. The line counts in the figure span total frequency ranges of 32 GHz (Sutton *et al.* 1985), 14.95 GHz (at frequencies ≤ 101 GHz from Cummins *et al.* 1986) and 18.6 GHz (Johansson *et al.* 1984, 1985).

It is clear that the $\log N$ - $\log T_A$ relationship for all 3 surveys is linear, which implies that $N \propto T_A^\alpha$, where $\alpha \approx -2$. Specific values of α from least square fits to the discrete points are shown in Figure 1.

From the power-law dependences of Figure 1 and the velocity line widths of Sgr B2 and Orion, a simple analysis reveals the sensitivity limits beyond which line overlap will become a serious problem. The results are shown in Table 2 where $T_A(\text{lim})$ signifies the antenna temperatures (for the same survey telescopes) at which the spectral lines become so dense that the probability is $\frac{1}{2}$ that a given line will overlap at least one other. The expected line densities shown in Table 2 indicate that 5 to 10 times more lines could, in principle, be detected in similar, more sensitive surveys before spectral overpopulation becomes a serious problem. However, in view of the fact that many of the lines detected in the three surveys arise from a few, heavy molecules, an increase by a factor of five in the number of

Recently, spectral line surveys in Sgr B2 (Cummins *et al.* 1986), Orion and IRC10216 (Johansson *et al.* 1984, Sutton *et al.* 1985) have been published which have implications for the future of line searches. A total of about 1200 lines were catalogued in these surveys, and all but 68 were identified and attributed to known species. The high percentage of identified lines casts a shadow on the earlier prediction of 100 known molecules before the end of the century. If a significant number of new molecules are to be found, the implications are that more sensitive surveys will be required.

lines detected does not imply a similar increase in the number of molecules.

TABLE 2. LINE CROWDING IN SGR B2 AND ORION

Source	Frequency Interval GHz	T_A (lim) K	Actual Line Density in Survey (GHz^{-1})	Expected Line Density at Sensitivity T_A (lim) (GHz^{-1})
Sgr B2	15 GHz between 71.9-101	0.013	12.7	60
Orion	18.6 GHz between 72.5-91.1	0.004	11.2	130
Orion	215-247	0.06	17.9	45

In my opinion, many of the new detections of large molecules will come from sensitive searches in TMC-1 for which sensitive, comprehensive surveys have not yet been published. In view of the small rotational constants of these molecules and the low T_{ex} which characterizes dark clouds, the most productive frequencies for TMC-1 searches are likely to be < 100 GHz. The frequency range from 25-50 GHz has been relatively neglected until recently and promises to be especially useful in heavy molecule studies. Even at the much studied frequencies above 70 GHz, little systematic, highly sensitive work has been done in TMC-1.

3. RECENT ASTRONOMICAL RESULTS

3.1. Observations

The last few years have seen an increase in the number of sources where larger molecules have been detected and studied. Benson and Myers (1983) have detected 10 new HC₅N sources in a survey of about 100 dark clouds with strong NH₃ (1,1) emission. Cernicharo *et al.* (1984) have discovered three condensations in Heiles' Cloud 2, in addition to TMC-1, where carbon chains up to HC₇N are detectable. These two studies bring the total number of known dark clouds with observed HC₅N to at least 21, with 13 of these, including all of the strong sources, in the Taurus-Auriga complex. Outside of the Taurus region Benson and Myers (1983) found relative abundances of HC₅N to be systematically lower by a factor of about 5.

TMC-1 continues to be the premiere dark cloud for difficult carbon chain detections. Bell and Matthews (1985) have succeeded in detecting the J=41-40 line of HC₁₁N at 13.9 GHz there, confirming the earlier observations in IRC10216 (Bell *et al.* 1982).

With the apparent detection of two lines of HC₉N in IRC10216 (Bell and Matthews, 1985, Matthews *et al.*, 1985), this circumstellar envelope joins TMC-1 as the only sources where all the known, linear

carbon chains have been detected. Other circumstellar envelopes have been shown to contain large molecules. HC_3N has been observed in three carbon star envelopes in addition to IRC10216, and HC_5N may have been detected in IRC40540 (Jewell and Snyder 1982, 1984). The presence of HC_3N in two bipolar nebulae (GL618 and GL2688) is now also established (Zuckerman *et al.* 1976, Jewell and Snyder, 1984), and there has been a possible detection in a comet (Hasegawa *et al.* 1984).

Another interesting development is the detection of HC_7N , HC_5N , C_4H and C_3N absorption lines against the supernova remnant Cas A (Bell *et al.*, 1983, 1986). The absorption appears at velocities corresponding to the local arm of the Galaxy (0 to -1.5 km s^{-1}) and to the Perseus arm (-35 to -60 km s^{-1}). A number of other, smaller molecules have been detected against Cas A including H_2CO (Goss *et al.*, 1984) and NH_3 (Batra *et al.*, 1984). However, the carbon-chain absorption appears to extend to more negative velocities in the Perseus arm than the smaller compounds, which indicates that they might not be entirely co-spatial. Bell *et al.* (1983) have suggested that the carbon chains could exist in a relatively diffuse component of the Perseus arm.

3.2. The Larger Molecules as Diagnostic Probes

A number of the relatively heavy molecules offer certain advantages as tools to investigate the physical conditions in molecular clouds. The symmetric tops, $\text{CH}_3\text{C}_2\text{H}$ and CH_3CN have energy levels structured so that multiple transitions corresponding to different K-ladders and excitation energies can be observed at nearly the same frequency. This eliminates systematic errors of pointing, calibration and source coupling which arise when different frequencies (and sometimes different telescopes) must be used to sample population distributions over excitation energy. In addition, their lines are strong enough to be readily detectable in a number of sources but are still optically thin. A number of authors have used these molecules to deduce molecular cloud temperatures. Two approaches have been used. In the simpler case it is assumed that the population distribution within a K-ladder is described by a single excitation temperature, T_r , which is the same for all ladders. Because radiative transitions between the K-ladders are forbidden, the total relative populations of different K-ladders are determined only by collisions and, hence, are functions only of kinetic temperature. Observations of two or more ΔJ transitions for $K=0,1,2,\dots$ can be used to estimate T_r and, hence, T_{kin} (e.g. Hollis *et al.* 1981, Kuiper *et al.* 1984).

A second, more rigorous approach is to carry out statistical equilibrium calculations which yield the distribution of population as functions of $n(\text{H}_2)$ and T_{kin} (e.g. Cummins *et al.*, 1983, Askne *et al.* 1984, Andersson *et al.* 1984). These calculations show that the T_r , T_{kin} two-temperature assumption can be significantly in error.

The recent detections of the $J=4-3$ HC_5N , and $N=1-0$ lines of C_3N and C_4H against Cas A (Bell *et al.* 1983, 1986) permit estimates of the densities where these molecules exist. Figure 2 shows calculated, line-centre brightness temperatures for the four carbon-chain transitions near 10 GHz observed by Bell *et al.* (1986) against Cas A. I

have computed T_b using statistical equilibrium calculations for $T_{kin} = 20K$, line widths of 15 km s^{-1} and a total molecular column density of 10^{13} cm^{-2} . The background brightness temperature of Cas A was taken to be 73K at 10 GHz, the beam-averaged value for the NRAO 43-m telescope where the lines were observed. Because the same column densities were used, the curves of Figure 2 depict the relative visibility, given equal abundances, of the four molecules. All of the transitions are optically thin against Cas A, so that T_b is proportional to column density.

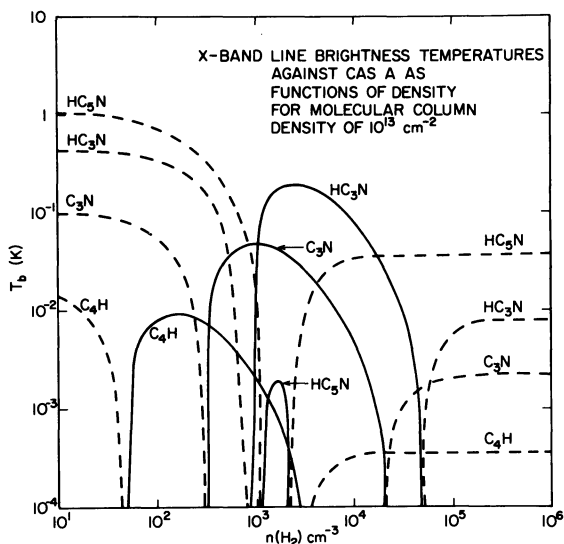


Fig. 2. Relative visibility curves for the $\Delta J=4-3$ HC_5N , $(\Delta J, \Delta F)=(1,2)-(1-1)$ HC_3N , $(\Delta N, \Delta J, \Delta F)=(1, 3/2, 5/2)-(0,1/2,3/2)$ C_3N and $(1,3/2,2)-(0,1/2,1)$ C_4H lines observed against Cas A. Solid curves indicate emission lines, dashed curves absorption lines.

possibility that these carbon chains exist in a low density component of the interstellar medium as suggested by Bell et al. (1983).

Another potentially important molecule is the recently discovered ring, C_3H_2 (Matthews and Irvine, 1985, Thaddeus et al., 1985b). To obtain a qualitative estimate of the potential of this molecule as a diagnostic tool and to tentatively identify key transitions, I have carried out statistical equilibrium calculations assuming optically thin lines for the ortho and para states. At present, collisional rate coefficients are unavailable, and I have used H_2O rates (Green 1980) scaled by a factor of 10 to compensate for the greater size and dipole moment of C_3H_2 . Both H_2O and C_3H_2 have b-type transitions.

Figure 2 shows that each of the four lines could appear either in absorption or emission against Cas A, depending upon $n(H_2)$. Emission occurs when $T_{ex} < 0$, which happens over a range of density that depends upon μ_B^2 which is different for each molecule (Avery, 1980). Of the four X-band transitions observed by Bell et al. (1983, 1986), HC_5N , C_4H and C_3N are in absorption, and the HC_3N results are ambiguous and require further observation.

The binary emission/absorption nature of spectral lines allows us to extract density information independent of relative line strengths, calibration errors, source coupling, atmospheric absorptions etc. From Figure 2 we note that the beam-averaged density, $n(H_2) > 2 \times 10^4 \text{ cm}^{-3}$ or $n(H_2) \lesssim 1 \times 10^2 \text{ cm}^{-3}$. For densities between these values, C_4H or C_3N would appear in emission. Unfortunately, this analysis, by itself, cannot confirm or rule out the

Among the strongest and most interesting of the C₃H₂ lines is the 1₁₀-1₀₁ ortho line at 18.343 GHz. It appears in many cold, dark clouds in emission and in rather exotic emission/absorption combinations against continuum sources (Matthews and Irvine 1985). Figure 3 shows calculated T_b ratios, for T_{kin} = 10K, of the 1₁₀-1₀₁ line at 18.3 GHz to some of the stronger, higher frequency C₃H₂ lines. The line frequencies are given in Table 3.

TABLE 3. FREQUENCIES OF C₃H₂ TRANSITIONS REFERRED TO IN FIGURES 3 AND 4.

Sym denotes whether the line belongs to the ortho (O) or para (P) state. (Thaddeus et al. 1985b).

Transition	Sym	Frequency (MHz)	Transition	Sym	Frequency (MHz)
1 ₁₀ -1 ₀₁	O	18343.143	2 ₀₂ -1 ₁₁	P	82093.559
2 ₂₀ -2 ₁₁	P	21587.395	2 ₁₂ -1 ₀₁	O	85338.905
3 ₂₁ -3 ₁₂	O	44104.787	3 ₁₂ -2 ₂₁	O	145089.635
2 ₁₁ -2 ₀₂	P	46755.621			

Figure 4 shows the results of similar calculations for ortho-para pairs of lines which are similar in frequency. The K-band pair, 1₁₀-1₀₁ and 2₂₀-2₁₁, appear to be the most sensitive indicators of density, whereas the 2₁₂-1₀₁/2₀₂-1₁₁ ratio appears unlikely to be a useful density discriminator.

Because of uncertainties in the collision rates and the assumption of optical thinness, Figures 3 and 4 should not be interpreted quantitatively, but the results do indicate the potential usefulness of C₃H₂ as a density discriminator.

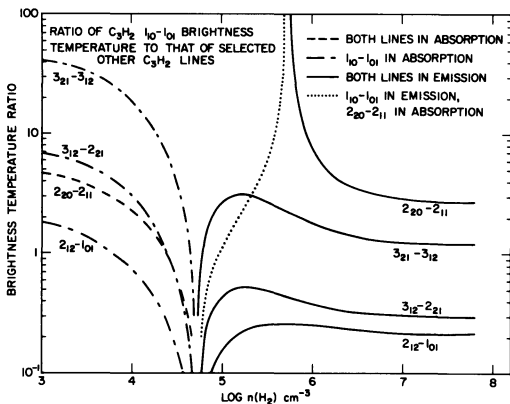


Fig. 3. Calculated ratio of line brightness temperatures for four pairs of C₃H₂ lines.

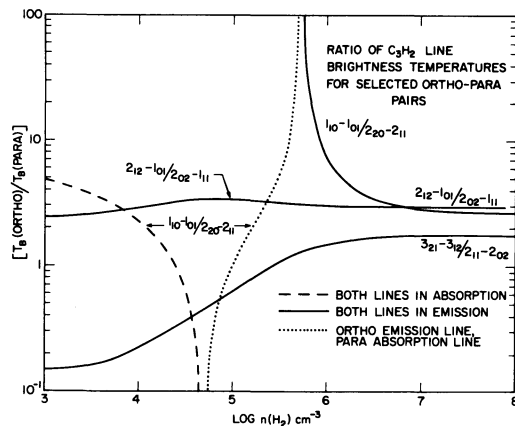


Fig. 4. Same as Figure 3 for three ortho-para pairs at similar frequencies.

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DISCUSSION

IRVINE: I will show later on some observational results that will confirm your predictions about the 18 and 21 GHz lines of $C_3H_2^+$. There is a density region where one appears in emission and the other in absorption in cold dark cloud.

AVERY: I am very pleased to hear that. My results had worried me having not gone to that high densities. This implies that at middle densities one is in emission and the other in absorption.

RAMADURAI: Has any line of CS or any other sulphide observed in Cas A or any other supernova remnant?

AVERY: To my knowledge, CS line has not been observed in the direction of Cas A.

PANDE: (1) If I correctly understood, you stated that molecules HC_3N and HC_5N have been identified in circumstellar envelopes. If so, which are these stars? What are their spectral and luminosity classes? Are they variable stars? I shall feel highly thankful if you can kindly provide the exact references. It may be a good idea to include such stars on regular spectrophotometric scanning (at our observatory) if possible.

AVERY: These are cool, carbon rich stars which have evolved off the

main sequence and are undergoing mass loss. They have extensive dusty envelopes in which these large molecules are observed. The prototypical and best-known is IRC+10°216, which is a variable star. The designations of the others I do not remember off hand, but I'll be happy to give you the references.

LEGER: Why carbon chains have been observed before carbon rings, if ever? Is it a matter of abundance or of ease of the detection?

AVERY: The carbon chains which have been detected have dipole moments which are larger than many of the 5 or 6 carbon rings which are most likely to be detected. The line strengths vary as the square of the dipole moment, so this is an important difference. Also, given comparable excitation conditions, the linear chains have smaller partition functions than the asymmetric top rings of similar molecular mass. Consequently, the spectral lines of the chains are more intense, molecule for molecule, than those of the rings. Taken together, these factors mean that rings are considerably more difficult to detect. A number of people have sought rings with 5 or 6 carbon atoms without success. My recollection is that current upper limits for rings in dense clouds are comparable to, or not significantly below, the abundances determined for chains of comparable mass.

GUELIN: In the case of rings with five to six heavy atoms, the abundance of rings is smaller than of chains. What one has to look for is a molecule with a high dipole moment like putting a nitrogen atom inside a ring, which is stable in the envelope.

K.K. GHOSH: Many complex molecules are detected in interstellar medium on grain surfaces (i.e. in Solid State). Why is it not possible to detect complex ring molecules in interstellar medium in gas-phase?

AVERY: Radio astronomy searches detect specific rotational lines of molecules which, as I understand it, are what has been attributed to grains. So searches for gas phase molecules can detect only one type of molecule at a time and generally require laboratory measurements of line frequencies, or else very precise structural information about the molecule. For many of the complex molecules suggested for grain surfaces, this information is unavailable. Also, the most stable complex molecules are apparently symmetric or nearly so, and have little or no dipole moment which makes them unobservable via the rotational transitions which are excited in the gas phase. Finally, and perhaps most important, the partition functions of complex molecules are large; this renders the individual lines of those, which do have dipole moments, very weak.