A Search for Interstellar Pyrimidine

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ABSTRACT

We have searched three hot molecular cores for submillimeter emission from the nucleic acid building-block pyrimidine. We obtain upper limits to the total pyrimidine (beam-averaged) column densities towards Sgr B2(N), Orion KL and W51 e1/e2 of 1.7 × 1014 cm−2, 2.4 × 1014 cm−2 and 3.4 × 1014 cm−2, respectively. The associated upper limits to the pyrimidine fractional abundances lie in the range (0.3–3) × 10−10. Implications of this result for interstellar organic chemistry, and for the prospects of detecting nitrogen heterocycles in general, are briefly discussed.

Key words: astrobiology – ISM: individual (Orion KL, Sgr B2(N), W51 e1/e2) – ISM: molecules – line: identification.

1 INTRODUCTION

Molecular clouds contain many organic molecules that are known to be important in biochemistry. Astronomical observations, particularly at radio wavelengths, allow us to determine the chemical composition and characteristics of this molecular inventory (e.g. Dickens et al. 2001; Charnley, Ehrenfreund & Kuan 2001). Following incorporation into protostellar disks and comets, these molecules, or their descendants, were probably the major source of volatile organic material available to the early Earth (Chyba et al. 1999). Studies of molecular cloud composition therefore enable us to quantitatively address the issue of the connection between interstellar chemistry, the organic composition of primitive Solar System material, and the origin, evolution and distribution of Life in the Galaxy (e.g. Ehrenfreund & Charnley 2001a; Ehrenfreund et al. 2002).

Many organics that are known, or strongly suspected, to be present in interstellar clouds, are fundamental components of the large organic macromolecules that are central to biochemistry. Examples of these are sugars and amino acids, the respective building blocks of polysaccharides and proteins. The simplest of these, glycolaldehyde and glycine, are both identified in the interstellar medium (Hollis et al. 2002; Kuan et al. 2003a).

Until recently, definitive detections of interstellar ring compounds have been scarce (e.g. ethylene oxide, Dickens et al. 1997) and previous searches for imidazole, cyaniform, pyrrole and pyrimidine were unsuccessful (Simon & Simon 1973; Myers, Thaddeus & Linke 1981, 1982). Recently we have tentatively detected the azaheterocyclic compounds 2H-azirine ([C2H3N]) and aziridine ([C2H5N]) at mm wavelengths (Kuan et al. 2003a; Charnley, Ehrenfreund & Kuan 2001). Two (different) tentative lines of Aziridine have also been claimed by Dickens et al. (2001). These observations suggest that biochemically important ring molecules may await detection. Of these potential discoveries, a key interstellar molecule for Astrobiology, comparable in importance to glycine, would be pyrimidine ([C4H6N2]), the unsubstituted ring analogue for three of the DNA and RNA bases: thymine, cytosine and uracil. Interstellar pyrimidine was unsuccessfully searched for at 46 GHz 30 years ago (Simon & Simon 1973). Evidence for various purines and pyrimidines in space, including pyrimidine, comes from the fact that they have been detected in meteoritic organic matter (Stoks & Schwartz 1983, 1984), and also may be components of Comet Halley’s CHON dust (Krueger, Korth & Kissel 1991).

The molecular composition of hot molecular cores is known to largely reflect the solid state chemistry that occurred on grain surfaces, prior to their deposition into the gas after protostellar dust heating (see Ehrenfreund & Charnley 2000). Reactions between HCN and VCN ([CH2CHCN] on the surface of dust grains has been suggested as a possible source of interstellar pyrimidine (Simon & Simon 1973). Observations of hot cores,
known to contain other rings believed to have formed on dust prior to evaporation, such as c-C$_2$H$_4$O (ethylene oxide) (Dickens et al. 1997; Nummelin et al. 1998) and c-C$_2$H$_3$N (Charnley, Ehrenfreund & Kuan 2001), do show the very high abundances of the HCN and VCN molecules required for formation of N-heterocycles (Ziurys & Turner 1991; Turner 1991; Nummelin & Bergman 1999). Alternatively, experiments indicate that energetic processing of ices (e.g. Allamandola, Bernstein & Sandford 1997) leads to an organic residue containing many polycyclic aromatic hydrocarbons (PAHs) (Greenberg et al. 2000). One may speculate that similar processing of interstellar ice analogues containing ammonia, molecular nitrogen, and hydrogen cyanide, may similarly produce N-heterocycles like pyrimidine.

Hence, searches of hot molecular cores, employing high-quality spectroscopic data, may offer the best chance of a pyrimidine detection. In this paper we report the results of searches of three massive star-forming regions for submillimeter emission from pyrimidine.

2 OBSERVATIONS

We carried out a program of submillimeter observations using the James Clerk Maxwell Telescope (JCMT)\(^1\) on Mauna Kea, Hawaii during Semester 01B over the period September 9–13, 2001. Additional measurements were made on November 23, December 1–2, and December 10, 2001. Our target list is given in Table 1. Column 4 gives the nominal LSR velocity of each source adopted for the search. The source list consists of three well-studied regions of massive star formation which are known to be rich in complex organic molecules (e.g. Blake et al. 1987; Miao et al. 1995; Remijan et al. 2002).

The heterodyne receivers used were the dual-channel B3 SIS mixers in the single-sideband mode (SSB) for the 345 GHz submillimeter band (315–373 GHz). The sideband rejection was ~13 dB (a factor of ~20). The SSB system temperatures in fair (medium) weather conditions ~ atmospheric opacity $\tau$ (225 GHz) between 0.08 and 0.12 were generally ~400 to 550 K. At 329.9 and 363.1 GHz, however, $T_{\text{sys}}$ as high as ~700 K and ~900 K were recorded, due to poor atmospheric transmission and higher receiver temperature. The half-power beamwidth (HPBW) of the telescope is ~14″ and the main-beam efficiency, $\eta_{\text{mb}}$, is 0.63. Data were obtained in the position-switching mode with offset 20° west in azimuth. Pointing and focus was checked regularly at a 2-hour interval. The resultant spectra are on the antenna temperature scale, $T_A^*$, which has been corrected from chopper wheels calibration for atmospheric transmission and losses associated with rearward scattering. Since the spatial extent of each emission source is not known, no main beam correction is applied. Further corrections for the forward scattering and spillover efficiency ($\eta_{\text{ss}} = 0.82$) convert the source antenna temperature, $T_A^*$, to the source brightness temperature, $T_B$.

By employing the Dutch Autocorrelation Spectrometer (DAS) backend with two subsystems and a bandwidth of 500 MHz for dual-polarity operation, we have a spectral resolution of 756 kHz and a channel spacing of 625 kHz. Typically an integration time of 3 to 4 hours was achieved. Velocity shifting of ±4.5 km s$^{-1}$ with respect to the nominal LSR velocity, about ±5 MHz with respect to the rest frequency in the 345 GHz band, was executed as a common practice during observations in order to neutralize the effect of possible low-level gain variations in the DAS and to identify potential interlopers from the image sideband. The JCMT data were reduced using the SPECX spectral line reduction package.

Observations of submillimeter high-frequency transitions yield smaller telescope beams, preferentially sample the warmer and denser regions, and help to avoid line confusion with interpolar emission from cooler envelope material along the line-of-sight. At hot core temperatures of 100 K or more, searches in the submillimeter regime are further favoured by the fact that the higher frequency transitions are expected to be intrinsically stronger.

Good agreement between observed line frequencies and those measured in the laboratory, for four or more spectral lines, are the minimum requirements for claiming identification of a new interstellar molecule. The rotational spectrum of pyrimidine has recently been measured over the spectral range 3–337 GHz, and the calculated dipole moment of pyrimidine is $\mu_{\text{rot}} = \mu_b = 2.39$ Debyes (Kisiel et al. 1999). Hence only $b$-type transitions are observable. Our astronomical search was based on the best candidate transitions of pyrimidine in spectral regions free from known spectral line contamination. Four bandheads made up of closely spaced high-$J$ transitions (at higher energy levels) plus 2 pairs of low-$J$ doublet lines were observed in a total of 6 different spectral bands. The observed transitions are listed in Table 2; transitions with line strengths smaller than 10.0 are not included. Column 1 lists the line number; each line may include multiple pyrimidine transitions which are blended into one unresolved, single spectral-line feature.

Figure 1 illustrates the predicted relative line intensity in an arbitrary scale of the pyrimidine spectra over a frequency range of 0 to 500 GHz at various rotational temperatures ($T_{\text{rot}} = 50$ K, the top panel; 100 K, the middle panel; and 200 K, the bottom panel). All possible spectral blends are accounted for in the simulation, which is made for a Gaussian lineshape with an assumed equivalent linewidth (FWHM) of 7 km s$^{-1}$. Note that the linewidth, when Doppler converted from velocity to frequency, increases with frequency, which is conducive to the formation of stronger blends at higher frequencies.

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**Table 1. Source List**

| Source          | R.A.(B1950) | Dec(B1950) | V$_{\text{LSR}}$ (km s$^{-1}$) |
|-----------------|------------|------------|-----------------------------|
| Orion KL        | 05° 32′ 47″.00 | −05° 24′ 30″.00 | 8.0                     |
| Sgr B2(N)       | 17° 44′ 10″.20 | −28° 21′ 15″.00 | 64.0                    |
| W51 e1/e2       | 19° 21′ 26″.30 | +14° 24′ 39″.00 | 60.0                    |
### Table 2. Pyrimidine transitions observed.

| Line | Rest Frequency (MHz) | Transition $J_{K_a,K_c} - J'_{K_a,K_c'}$ | $S_{d}^I$ | $E_{d}^I$ (cm$^{-1}$) |
|------|---------------------|---------------------------------|--------|-----------------|
| 1    | 329961.004          | 531.53 - 520.52                 | 52.48  | 288.87          |
| 2    | 336125.535          | 541.54 - 530.53                 | 53.48  | 299.88          |
| 3    | 338017.612          | 2727.1 - 2636.0                 | 24.97  | 146.26          |

### Table 2. (continued).

| Line | Rest Frequency (MHz) | Transition $J_{K_a,K_c} - J'_{K_a,K_c'}$ | $S_{d}^I$ | $E_{d}^I$ (cm$^{-1}$) |
|------|---------------------|---------------------------------|--------|-----------------|
| 4    | 342289.902          | 551.55 - 540.54                 | 54.48  | 311.09          |
| 5    | 348453.267          | 4412.32 - 4312.31               | 31.89  | 290.41          |

A Search for Interstellar Pyrimidine
Table 2. (continued).

| Line | Rest Frequency (MHz) | Transition | \( S_{ul} \) | \( E_{l}^1 \) (cm\(^{-1}\)) |
|------|----------------------|------------|-------------|--------------------------|
| 348458.771 | 4115.26 – 3406.25 | 26.30 | 273.12 |
| 348458.771 | 4116.26 – 3406.25 | 26.30 | 273.12 |
| 6 | 363098.770 | 2\(28,1 - 28,0\) | 26.97 | 169.23 |
| 363099.057 | 2\(29,0 - 28,28,1\) | 26.97 | 169.23 |

* Line Strength.
† Lower energy level.

The strongest lines arise from blends of many transitions in high-J bandheads. These bandheads become more compact and have more lines at higher frequencies. On the other hand, following Boltzmann population distribution, intensities of individual transitions forming the spectral blends, or bands, maximize at appreciably lower frequencies (henceforth the Boltzmann maxima) than the maxima apparent in the spectral profiles in Figure 1. At \( T_{rot} = 100 \) K, for example, the strongest individual band line, i.e., the Boltzmann maximum, though not obvious in the spectrum (the middle panel of Figure 1), is near 195 GHz. Nevertheless, because at such frequencies the band compression effect becomes dominant and compensates for the intensity decrease of transitions at frequencies beyond the Boltzmann maximum, the overall band profile reaches an intensity maximum at the much higher frequency of \( \sim 300 \) GHz. Similarly at \( T_{rot} = 50 \) K and 200K, the Boltzmann maxima are at \( \sim 135 \) GHz and \( \sim 285 \) GHz, respectively, while at higher frequencies where band compression dominates, the apparent maximum intensity features for a linewidth of 7 km s\(^{-1}\) are predicted to be at \( \sim 180 \) GHz and \( \sim 335 \) GHz, accordingly. The 324 GHz spectral blend, which protrudes above the general profile and is seen in the middle and bottom panels of Figure 1, arises from accidental addition of some non-band lines to the spectral blend. The prevailing spectral features at 100 K and 200 K are compact high-J R-type bandheads of the type observed in our search (see Table 2). For an excitation temperature near 100 K, it is clear that searches in the spectral region near 1-mm (300 GHz) should provide the best chance for a detection.

3 RESULTS

Table 3 lists the important measured physical parameters of all the pyrimidine lines observed. The pyrimidine lines are separated into three sources according to the actual observations (Column 1). Column 2 gives the line numbers, which correspond to the line numbers shown in Column 1 of Table 2. Most of the pyrimidine lines are only observed in one or two sources but not in all. The fact that most of the data are fragmentary is mainly due to scheduling difficulties at the JCMT, leading to limited or uncertain availability of observing time for different sources.

In total, 6 different pyrimidine lines were observed toward the 3 target sources. The peak antenna temperature of the spectral line observed, i.e. the \( T_{A}^\star \) upper limit for a non-detection, is given in Column 3 in mK; “rms” indicates the 1-\sigma noise level per channel of the spectrum, where \( \sigma \equiv T_{A}^\star (\text{rms}) \).

Column 4 lists the upper limits of the total column density \( N_{tot} \). \( T_{A}^\star \), hence also \( N_{tot} \), are not listed if the target pyrimidine line is completely overwhelmed by nearby strong interlopers. Self-explanatory comments for each spectral line observed in each source are given, when necessary, in Column 5. Interlopers, which are largely blended with (denoted as “(B)”) and smeared out the target pyrimidine lines or partially blended (denoted as “(PB)”) with the candidate lines, are also included in Column 5 in the format molecule/transition(MHz).

Assuming that the pyrimidine lines are optically thin, in LTE, and that the rotational excitation temperature, \( T_{rot} \), is much higher than the background brightness temperature, the beam-averaged total column density of c-C\(_4\)H\(_4\)N\(_2\) can be written as:

\[
N_{tot}(\text{cm}^{-2}) = (1.669 \times 10^{17}) \left( \frac{W_{K}Q_{rot}}{\nu} \right) \left( \sum \left( \frac{S_{ul} \mu_{b}^2}{\exp \left( \frac{E_{u}}{T_{rot}} \right)} \right) \right)^{-1}, \quad (1)
\]

where

\[
W_{K} = \int T_{R}^{\star} dv = \int \frac{T_{A}^{\star}}{\eta_{ss}} dv \quad (2)
\]

in (K km s\(^{-1}\)) is the integrated intensity of the spectral line either from a single pyrimidine transition, or from unresolved multi-transitions such as in a bandhead. The rest frequency (MHz) is \( \nu \), \( \mu_{b} \) the permanent b-dipole moment in Debye, \( S_{ul} \) the line strength, \( E_{u} \) the upper energy level in K and \( Q_{rot} \) the rotational partition function. All pyrimidine lines reported here contain more than one pyrimidine transition, a summation of \( (S_{ul}\mu_{b}^2)/[\exp(E_{u}/T_{rot})] \) over all relevant transitions was thus applied. In addition, a rotational temperature \( T_{rot} = 100 \) K was assumed for all transitions. In the case of a non-detection, for an interloper-blended pyrimidine line, \( T_{R}^\star \) is obtained from the \( T_{A}^\star \) upper limit listed in Column 3 of Table 3 corrected for \( \eta_{ss} \). If the equivalent linewidth could not be unambiguously defined from the observed spectrum, the column density limits were evaluated using a representative value of the equivalent linewidth; a value of \( \Delta v \approx 10 \) km s\(^{-1}\) was adopted for all three target sources.

Sample pyrimidine spectra are shown in Figures 2 and 3. With a limited number of spectral lines observed in each source, our submillimeter-wave search for pyrimidine did not yield a definite detection of pyrimidine in either Sgr B2(N) or Orion KL. By averaging over all observed lines measurable in each source (see Table 3), the inferred upper limits on the total column density in Sgr B2 and Orion are \( 1.7 \times 10^{14} \) cm\(^{-2}\) and \( 2.4 \times 10^{14} \) cm\(^{-2}\), respectively.

In the case of W51 e1/e2, there is a spectral feature at \( \sim 57 \) km s\(^{-1}\) with respect to the rest frequency adopted for the observation (see the spectrum shown in the lower panel of Figure 3). This feature coincides precisely with the J \( = 55 \) band-head of pyrimidine between 342289.9 and 342297.4 MHz (Line 4), containing 32 transitions at \( V_{LSR} = 59.0 \) km s\(^{-1}\). An unidentified U-line at 342290.0 MHz was previously reported in a line survey of Orion KL [Schilke. et al. 1997]. Unfortunately we did not observe either Orion or Sgr B2 at this particular frequency.

The total pyrimidine column density in W51 e1/e2 derived solely from Line 4 is \( 4.8 \times 10^{14} \) cm\(^{-2}\); the averaged upper limit to the column density from all 3 lines available in
W51 is $N_{\text{tot}} \leq 3.4 \times 10^{14} \text{ cm}^{-2}$. It is interesting to note that the pyrimidine column density computed only from Line 4 (assuming $T_{\text{rot}} = 100 \text{ K}$) is just slightly higher (within a factor of 2) than the upper limits deduced from the two other, interloper-contaminated, lines. It appears that if Line 4 of W51 is truly a detection, we may have expected to detect another bandhead, Line 5, whose transitions are at higher frequencies. It could be argued that the fact that we failed to detect Line 5 makes the identification of Line 4 less likely.

However, Line 5 is expected to be much weaker than Line 4 at an excitation temperature lower than 100 K, and so, in the absence of accurately measured pyrimidine rotational temperatures for these sources, we cannot entirely rule out a true detection in W51.

Finally, the submillimeter transitions targeted in this search possess rather high excitation levels, and so any observable pyrimidine emission would mostly originate from regions of high temperature and/or density, if radiative ex-
cition is negligible. As a result, these excited pyrimidine molecules would have been located in regions fairly close to protostellar hot cores within molecular clouds. It is therefore probable that the real source size is smaller than the telescope beam (~14″); it is more likely to be the case particularly for distant sources such as Sgr B2(N) and W51 e1/e2. Hence, one should note that our observations would suffer from beam dilution if the source extent is indeed smaller than the beam. The derived upper limits on the column densities, based on the assumption that the source fills the beam, could consequently be underestimated.

The upper limits of fractional abundance of pyrimidine with respect to molecular hydrogen, \(X(c - C_4H_2N_2) = \frac{N_{tot}(c - C_4H_2N_2) \div N_{tot}(H_2)}{N_{tot}(c - C_4H_2N_2) \div N_{tot}(H_2)}\), may also be deduced. Molecular hydrogen column densities inferred from single-dish observations with beam sizes similar to the JCMT are employed, in order to determine the beam-averaged abundances more accurately. The \(H_2\) column densities adopted are: \(5 \times 10^{24}\) cm\(^{-2}\) for Sgr B2(N) (Nummelin et al. 2000), \(8 \times 10^{23}\) cm\(^{-2}\) for Orion KL (Sutton et al. 1995), and \(1 \times 10^{24}\) cm\(^{-2}\) for W51 e1/e2 (Jaffe, Becklin & Hildebrand 1984). The upper limits of pyrimidine fractional abundances thus estimated are \(X(c - C_4H_2N_2) \leq 3.4 \times 10^{-11}\) for Sgr B2, \(3.0 \times 10^{-10}\) for Orion, and \(3.4 \times 10^{-10}\) for W51.

### 4 DISCUSSION

Our submillimeter search for the nucleic acid building-block pyrimidine has not been successful. We have perhaps detected one-line in a single source but this needs to be confirmed. The negative result therefore cannot be considered definitive and searches at lower frequencies may yet detect pyrimidine in hot cores. The low inferred pyrimidine abundance limits may simply reflect the fact that pyrimidine is generally of low abundance in massive star-forming cores, and hence it is difficult to populate the high-J energy levels \((J \leq 56\) in our study) which are observable at submillimeter wavelengths. On the other hand, it could also imply that either pyrimidine is not evaporated efficiently from dust grains at hot core temperatures, or is destroyed easily in the warmest regions in hot cores sampled by submillimeter observations.

It would appear that the best chance for an astronomi-
A Search for Interstellar Pyrimidine

Figure 2. Sample pyrimidine spectra at 338.0 GHz (Line 3; 27,1-26,0 and 27,0-26,1) of Sgr B2(N) (top) and of W51 e1/e2 (bottom). The two vertical lines mark the two expected transitions of pyrimidine. The abscissae give the LSR velocities with respect to the rest frequencies adopted for the observations at the nominal LSR velocities 64.0 km s\(^{-1}\) for Sgr B2(N) and 60.0 km s\(^{-1}\) for W51 e1/e2.

Figure 3. Sample pyrimidine spectra at the 336.1 GHz band head (Line 2; with J \(\leq 54\)) of Orion KL (top), and at the 342.3 GHz band head (Line 4; with J \(\leq 55\)) of W51 e1/e2 (bottom). The two verticals enclose the 336.1 GHz band head (top) and the 342 GHz band head (bottom). A tentative detection of pyrimidine in W51 e1/e2 is visible in the bottom panel. The assumed LSR velocities for the rest frequencies adopted for the observations are 8.0 and 60.0 km s\(^{-1}\) for Orion KL and W51 e1/e2, respectively.

Experiments appear to indicate that any circumstellar pyrimidine would be easily destroyed by UV photons, or cosmic ray particles, soon after delivery to the interstellar medium. In this case, any interstellar pyrimidine would have to be produced in dark molecular clouds.

5 CONCLUSION

To strengthen the role of prebiotic interstellar matter in Astrobiology, we have searched for 6 pyrimidine lines in three massive star-forming regions: Sgr B2(N), Orion KL and W51 e1/e2. Our search was unsuccessful and did not yield a conclusive result, with only one potential single-line detection. The abundance limits inferred are \(\leq 3.4 \times 10^{-11}\) for Sgr B2, 3.0 \(\times 10^{-10}\) for Orion, and 3.4 \(\times 10^{-10}\) for W51. Our negative result may simply reflect the fact that interstellar pyrimidine is of low abundance. Sources that are ongoing sites of carbon dust formation, such as the C-rich envelopes of AGB and post-AGB stars, probably present the best opportunity for detecting pyrimidine and other nitrogen heterocycles.

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