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CHEMICAL TIMESCALE OF DARK CLOUD CORES ESTIMATED FROM DEUTERIUM FRACTIONATION

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ABSTRACT

The chemical evolutionary age of dark cloud cores is estimated from the degree of deuterium fractionation, on the assumption that the chemical model used is reasonable. The method is applied to dark cloud cores along the TMC-1 ridge, using the abundance ratios between DCO^+ and $H^{13}CO^+$ combined with the new standard model network of gas-phase chemical reactions. The difference in deuterium fractionation between the ammonia peak and the cyanopolyyne peak, though its error transferred from the observed data is relatively large, is explained by a time difference in the evolutionary age of more than 10^5 yr, or by a small change in the depletion factor of carbon and oxygen, which also indicates the degree of core evolution. The method of determining the evolutionary age of dark cloud cores is somewhat free from the details of chemical reaction not directly related to deuterium fractionation. The present result is compared with those from other methods.

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

1. INTRODUCTION

More than 120 molecular species have been detected and identified in space. The mechanism of their production and destruction has been proposed, and chemical models have been developed to explain and predict the existence and abundance of various interstellar molecules (for example, Herbst & Leung 1989, 1990; Lee, Bettens, & Herbst 1996). The predicted abundances of some main species are in agreement with observed values, but for many other molecules, especially heavy atom-bearing molecules and large polyatomic molecules, there is occasionally a large difference between the predicted and observed abundances. Revisions of chemical models have been made by including the grain-surface chemistry (Hasegawa & Herbst 1993), neutral-neutral reactions (Herbst et al. 1994), and grain-surface molecular depletion and desorption (Bergin, Langer, & Goldsmith 1995). Using a time-dependent chemical model, Suzuki et al. (1992) proposed that the relative magnitude of molecular column densities can be interpreted as an effect of chemical evolution of dark clouds, and the ratio [CCS]/[NH_3] may serve as an indicator of cloud evolution.

Hirahara et al. (1992) carried out high-resolution mapping observations of carbon-chain molecules and NH_3 along the ridge of TMC-1 and proposed that the difference in molecular distribution of CCS and NH_3 between the cyanopolyyne peak and the ammonia peak can be ascribed to their different stages of chemical evolution. On the basis of a one-dimensional hydrodynamical simulation of a self-gravitating gas cloud, Hanawa, Yamamoto, & Hirahara (1994) proposed a sequential core collapse along the ridge of TMC-1. Howe, Taylor, & Williams (1996) presented a chemical model for collapsing cloud cores and suggested that the chemical evolutionary age, consistent with the column densities of carbon-chain molecules and NH_3, could be deduced for each core in TMC-1. Pratap et al. (1997) carried out intensive mapping observations of several molecular species along the ridge of TMC-1. They found that the observed abundance gradients could be explained by a small variation in the chemical timescale from 1.2×10^5 to 1.8×10^5 yr, by a change in density by a factor of 2 along the ridge, or by a variation in the C/O ratio from 0.4 to 0.5 along the ridge. Recently, Markwick, Millar, & Charnley (2000) presented an interesting explanation to this problem: the density gradients along the ridge were produced following the explosive desorption of photolyzed ices, induced by MHD waves as they propagate inside the cloud.

Several deuterated species are detected toward TMC-1 (Langer et al. 1980; MacLeod, Avery, & Broten 1981; Schloerb et al. 1981). In 1982, Guélin and his collaborators carried out mapping observations of five isotopic species of HCO^+ toward several dense molecular clouds including TMC-1. They found that HCO^+ is distributed in a wide region of the TMC-1 ridge, showing its peak position at about $1'.5$ northwest of the ammonia peak, whereas the peak position of DCO^+ coincides with the ammonia peak. Later, Butner, Lada, & Loren (1995) conducted a survey observation of DCO^+ and $H^{13}CO^+$ in a sample of low-mass cores including the ammonia peak of TMC-1 and determined the [DCO^+]/[HCO^+] fractionation ratio to be 0.027 for the ammonia peak.

Bell et al. (1988) detected c-C_3HD toward several molecular clouds. They obtained very high abundance ratios of c-C_3HD to c-C_3H_2 in the TMC-1 region: 0.08 at the cyanopolyyne peak, 0.12 in the region of the ammonia peak, and 0.16 in the region of the HCO^+ peak. This high fractionation of the deuterated species necessitated a reconsideration of the chemical formation routes to c-C_3H_2 and c-C_3HD. Howe & Millar (1993) presented two chemical models that

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give the degree of the deuterium fractionation for c-C_3H_2 to be ~0.15.

In 1994, Howe et al. carried out extensive high-resolution mapping observations of DC_3N and HC_3N toward several molecular clouds, especially a few positions along the TMC-1 ridge centered at the cyanopolyyne peak. They obtained a high deuterium fractionation of a few to 10 percent, which was explained by linking the deuteration of HC_3N to that of c-C_3H_2. Recently, we carried out a spectral line survey toward the cyanopolyyne peak of TMC-1 by using the Nobeyama 45 m radio telescope (Kaifu et al. 2002) and obtained relative abundances for four deuterated species: DC_3N, DC_5N, HDCS, and c-C_3HD. Very recently, Tiné et al. (2000) detected NH_2D and N_2D^+ toward the ammonia core and reported abundance ratios of 0.02 and 0.08 with respect to NH_3 and N_2H^+, respectively. Hirota, Ikeda, & Yamamoto (2001) carried out mapping observations of DNC and $HN^{13}C$ toward TMC-1 and found [DNC]/[HNC] to be 0.028 and 0.042 toward the cyanopolyyne peak and the ammonia peak, respectively. The observed relative abundances of the deuterated species in TMC-1 are summarized in Table 1.

The high chemical fractionation of the deuterated species occurs in the gas phase at low temperatures (Watson 1976, 1977), and the degree of fractionation depends on the time elapsed (Millar, Bennett, & Herbst 1989; Howe & Millar 1993). We suggest in this paper that the degree of chemical fractionation of selected molecular species can be used to estimate the chemical evolutionary age of dark cloud cores. In a similar but more comprehensive vein, Roberts & Millar (2000a) published a paper on the modeling of deuterium chemistry. They presented new models for investigating deuterium fractionation in molecular clouds with a wide range of physical parameters, including density, temperature, elemental abundances, and the freeze-out of molecules on to dust grains, and they showed how fractionation can be used to probe the history of interstellar matter.

TABLE 1

DEUTERIUM FRACTIONATION IN TMC-1[a]

Molecule	CP Peak[b]	NH_3 Peak[c]	HCO^+ Peak[d]
$R(HCO^+)$	1.7,[e] 1.2[f]	2.7[g]	
$R(C_3H_2)$	8,[h] 7.5,[i] 4.75[f]	12,[h] 3–6[j]	16[h]
$R(HC_3N)$	5–6,[k] 3–6,[l] 2,[i] 1.45[f]		
$R(HC_5N)$	1.2,[m] 1.6,[n] 3[i]		
$R(C_2H)$	4.8(15)[f]		
$R(C_4H)$	0.43[o]		
$R(H_2CO)$.........		5.9[f]	
$R(H_2CS)$	2[p]		
$R(NH_3)$...........	0.085[f]	2,[q] 4.3[f]	
$R(N_2H^+)$	0.64(13)[f]	8,[q] 0.98[f]	
$R(HCN)$..........	1.10 (46)[f]		
$R(HNC)$..........	2.8,[r] 1.6[f]	4.2[r]	
$R(CH_3CCH)$...	5.1–6.1[s]		
$R(CH_3OH)$	2.65[f]		

[a] $R(XH) = [N(XD)]/[N(XH)] \times 100$.

[b] Cyanopolyyne peak, $\alpha = 4^{\mathrm{h}}38^{\mathrm{m}}38^{\mathrm{s}}.6$, $\delta = 25°35'45''$ (1950.0).

[c] $\alpha = 4^{\mathrm{h}}38^{\mathrm{m}}19^{\mathrm{s}}.0$, $\delta = 25°42'30''$ (1950.0).

[d] $\alpha = 4^{\mathrm{h}}38^{\mathrm{m}}12^{\mathrm{s}}.0$, $\delta = 25°44'00''$ (1950.0).

[e] Caselli et al. 1998.

[f] Turner 2001. Values in parentheses denote the errors quoted and apply to the last digits of the relative abundance ratios.

[g] Butner et al. 1995.

[h] Bell et al. 1988.

[i] Kaifu et al. 2001.

[j] Gerin et al. 1987.

[k] Langer et al. 1980.

[l] Howe et al. 1994.

[m] MacLeod et al. 1981.

[n] Schloerb et al. 1981.

[o] Turner 1989.

[p] Minowa et al. 1997.

[q] Tiné et al. 2000.

[r] Hirota et al. 2001.

[s] Gerin et al. 1992, referred to CH_2DCCH.

2. CHEMICAL EVOLUTIONARY AGE OF CORES IN TMC-1

Only three deuterated species, DCO^+, c-C_3HD, and DNC, have been studied so far at both the cyanopolyyne peak and the ammonia peak of the TMC-1 ridge, where physical conditions are relatively well known (Table 1). HCO^+ is one of the most fundamental molecular ions in interstellar gaseous chemistry, and DCO^+ is a primary species in interstellar deuterium fractionation, to which the deuterium atom is transferred from highly fractionated H_2D^+ at low temperatures (~10 K). Both HCO^+ and DCO^+ have been detected and studied in a wide variety of molecular clouds, from dark clouds to massive star-forming regions.

As noted above, c-C_3HD shows a very high relative abundance as well as a large change along the TMC-1 ridge. However, Turner (2001) recently reported that his analyses of C_3H_2 and C_3HD disagreed with the earlier analyses, and much lower fractionation values were derived for TMC-1 cores. The new analyses are consistent with a new prediction that the key reaction to produce the C_3HD species,

$$C_3H_3^+ + HD \rightarrow C_3H_2D^+ + H_2 \; ,$$

has a potential barrier of about 4 eV and does not contribute at all to deuterium fractionation at temperatures less than 10 K (Talbi & Herbst 2001).

Although the relative abundance between DNC and HNC was observationally determined with relatively good accuracy, the production and fractionation of these species have an ambiguity, and the relative abundance is not suitable for quantitative analysis, because the branching ratio in dissociative recombination of $HCND^+$ is not known. Furthermore, several chemical model calculations so far presented show a large hump of the DNC abundance around 10^5 to 10^6 ages (for an example, Howe & Millar 1993). This means that a given relative abundance of DNC corresponds to two separate years for the limited period.

The relative abundance of DCO^+ at the cyanopolyyne peak was reported to be 1.7% ± 0.3% by Caselli et al. (1998) using the data of DCO^+ and $H^{13}CO^+$ by Guélin, Langer, & Wilson (1982), and that at the ammonia peak to be 2.7% ± 0.5% using the data by Butner et al. (1995). Caselli et al. (1998) analyzed the abundance ratios $R_D = [DCO^+]/[HCO^+]$ and $R_H = [HCO^+]/[CO]$ of several dark cloud cores so as to determine the electron fraction, $\chi(e)$, the cosmic-ray ionization rate, ζ, and the C/O depletion degree, f_D. The depletion factor, f_D, is defined such that a fraction $1/f_D$ of the O and C in the standard "low metal" abundances remains in the gas phase. Their analysis is based on a simple analytical chemical model of deuterium fractionation and the steady state abundances derived from a detailed pseudo–time-dependent chemical model. They derived physical conditions at both the cyanopolyyne peak and the ammonia peak, but the steady state physical condition at the cyanopolyyne peak is inappropriate, as they pointed out, because their steady state analysis cannot reproduce the abundances of complex molecules observed toward TMC-1(CP). Pratap et al. (1997) reported that the kinetic temperature along the TMC-1 ridge is derived to be 10 K from CH_3CCH and NH_3 lines, which is almost consistent with the temperature derived from ^{12}CO and ^{13}CO lines. They also derived the molecular hydrogen density, $n(H_2)$, to be 8.3×10^4 and 6.2×10^4 cm^{-3} at the CP and NH_3 peaks, respectively, from the analysis of HC_3N lines.

The degree of deuterium fractionation depends on the physical conditions of the cloud core where molecules evolve. It is essential to use plausible physical conditions for a quantitative analysis of deuterium fractionation. As noted above, Caselli et al. (1998) carried out an extensive and intensive analysis of physical conditions for many dark clouds, varying the ionization rate. We think the ionization rate does not change far from the standard value, 1.3×10^{-17} s^{-1}, in such dark cloud cores. No ultraviolet radiation sources along the TMC-1 ridge are known, and the cosmic-ray ionization rate can be safely assumed to be the standard value. If the molecular hydrogen densities are assumed to be those obtained by Pratap et al. (1997) for the CP and NH_3 cores, the simple analytical chemical model used by Caselli et al. (1998), that is, an analysis using equations (3) and (4) of their paper, gives f_D to be about 1 based on the observed R_D and R_H values toward both peaks. Therefore, it is reasonable to choose physical conditions for both cores to be $T_K = 10$ K, $n(H_2) = 7 \times 10^4$ cm^{-3}, $\zeta = 1.3 \times 10^{-17}$ s^{-1}, and $f_D = 1$, physically static conditions, which are used to calculate the relative abundance of $[DCO^+]/[HCO^+]$ by employing the new standard model network of gas-phase chemical reactions (Terzieva & Herbst 1998). The network has been extended to include monodeuterated species of hydrogen-bearing molecules (Millar et al. 1989; Aikawa & Herbst 1999). For normal exothermic reactions and dissociative recombination reactions, the total rate coefficient is assumed to be unchanged for deuterated species and statistical branching ratios are also assumed. $HCO^+(DCO^+)$ is mainly produced by the reaction of CO with $H_3^+(H_2D^+)$ and destroyed by the dissociative recombination reaction with electrons (Dalgarno & Lepp 1984). In total, 639 molecular species and 10,830 chemical reactions are included in the network. The model is pseudo–time-dependent, and we assumed an initial state where all elements except hydrogen are atomic with "low metal" abundances of $\chi(O) = 1.8 \times 10^{-4}$, $\chi(C) = 7.3 \times 10^{-5}$, $\chi(S^+) = 8 \times 10^{-8}$, $\chi(Si^+) = 8 \times 10^{-9}$, $\chi(Mg^+) = 7 \times 10^{-9}$, $\chi(Fe^+) = 3 \times 10^{-9}$, and $\chi(Na^+) = 2 \times 10^{-9}$. All deuterium was assumed to be present as HD in the initial conditions and its fractional abundance to be 3.0×10^{-5} (Millar et al. 2000). The calculated relative abundance ratio $R(HCO^+) = 100 \times R_D$ is shown by the curve (a) in Figure 1 as a function of time, where the observed relative abundances with error bars are also indicated at the right-hand side of the figure.

As Figure 1 shows, there is a difference of about 0.5% in the relative abundance between the observed value toward the ammonia peak and the predicted value at steady state, though the observed values show relatively large errors. If we assume that the observed value toward the ammonia peak corresponds to the steady state value, then, if the 0.5% difference is time-independent, that of the cyanopolyyne peak is deduced to correspond to the relative abundance at 1.6×10^4 yr. Since steady state pertains at $\gtrsim 1 \times 10^5$ yr, it may be concluded that the difference between the evolutionary ages between the ammonia peak and the cyanopolyyne peak is probably more than 10^5 yr.

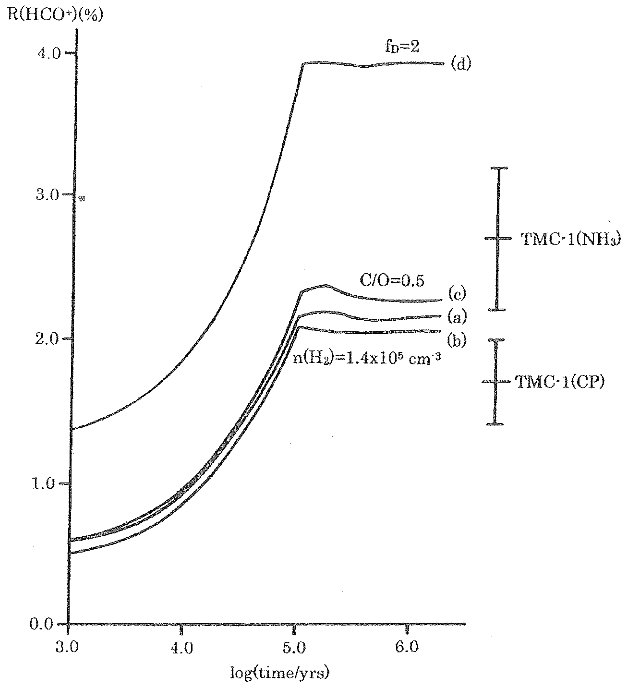

FIG. 1.—The calculated relative abundance ratio between DCO^+ and HCO^+, $R(HCO^+)$ [$= 100 \times R_D$ (Caselli et al. 1998)], using the new standard model network with $\zeta = 1.3 \times 10^{-17}$ s^{-1}, and "low metal" abundances of $\chi(O) = 1.8 \times 10^{-4}$, $\chi(C) = 7.3 \times 10^{-5}$, $\chi(S^+) = 8 \times 10^{-8}$, $\chi(Si^+) = 8 \times 10^{-9}$, $\chi(Mg^+) = 7 \times 10^{-9}$, $\chi(Fe^+) = 3 \times 10^{-9}$, and $\chi(Na^+) = 2 \times 10^{-9}$ at 10 K. Curve (a): $n(H_2) = 7 \times 10^4$ cm^{-3}, $f_D = 1$, and $C/O = 0.4$; curve (b) $n(H_2) = 1.4 \times 10^5$ cm^{-3}, $f_D = 1$, and $C/O = 0.4$; curve (c) $C/O = 0.5$, $n(H_2) = 7 \times 10^4$ cm^{-3} and $f_D = 1$, and curve (d) $f_D = 2$, $n(H_2) = 7 \times 10^4$ cm^{-3}, and $C/O = 0.4$. The observed values at the cyanopolyyne peak and the ammonia peak are indicated with error bars at the right-hand side.

3. DISCUSSION

Suzuki et al. (1992) showed that carbon-chain molecules including CCS are abundant in the early stages of chemical evolution, whereas NH_3 is abundant in the later stages, on the basis of pseudo–time-dependent chemical reaction network simulations. The conclusion suggests that the cyanopolyyne peak of the TMC-1 ridge is in an earlier evolutionary stage than the ammonia peak. Although it is qualitative, this suggestion was reinforced by Hirahara et al. (1992), who carried out high-resolution mapping observations of several molecules along the TMC-1 ridge and showed a gradual change in the ratio between the column densities of NH_3 and carbon-chain molecules along the ridge. The sense of the present study, that the ammonia peak is older than the cyanopolyyne peak, is in qualitative agreement with the standard gas-phase models which show that CCS peaks at early times and ammonia peaks at steady state.

From their intensive mapping observations along the TMC-1 ridge, Pratap et al. (1997) concluded that the observed abundance gradients along the ridge can be explained by a small variation in the chemical timescale from 1.2×10^5 to 1.8×10^5 yr. Their conclusion is based on the relative abundance between CS and SO along the ridge, which is suggested to be a very sensitive indicator of the evolutionary state of the cloud (Bergin et al. 1997), according to time-dependent chemical model calculations (Bergin et al. 1995; Bergin, Snell, & Goldsmith 1996). They also suggested alternatively that the observed gradients can be produced by varying the density by a factor of 2 or the C/O ratio from 0.4 to 0.5 along the ridge.

We investigated the dependence of the abundance ratio, R_D, on physical parameters, using the new standard model. A change in the density by a factor of 2 gives only a change of less than 0.1% in the abundance ratio, as indicated in the curve (b) of Figure 1. A change of the C/O ratio from 0.4 to 0.5 produces only a similar small change of less than 0.2%, as illustrated in the curve (c) of Figure 1. Pratap et al. (1997) suggest that the high C/O ratio reproduces high column densities of carbon-bearing species at the CP core, whereas the low C/O value prefers oxygen-bearing species at the NH_3 core. However, this figure shows that the change in R_D as C/O varies from 0.4 to 0.5 is contrary to the direction of change of observed values at the CP to the NH_3 cores. Similarly, a change in the ionization rate or the metal abundance by a factor of 2, which is exemplified in the analysis of Caselli et al. (1998) for the two peaks of TMC-1, also gives only a small change of less than 0.2% in the abundance ratio, R_D, from 10^4 yr to the steady state. An increase of the depletion factor of carbon and oxygen raises the relative abundance to a higher value. For example, the relative abundance of DCO^+ increases by 1.0%–1.5% when f_D is increased from 1 to 2, as shown in the curve (d) of Figure 1. The observed R_D variation between the peaks can therefore be explained by introducing an f_D value between 1 and 2. However, f_D may also change during the physical evolution of the cloud core; the more cloud evolves, the higher f_D. This means that f_D is an indicator of the evolutionary state of the core, as pointed out by Roberts & Millar (2000a, 2000b). A temporal increase in f_D thus raises calculated R_D values at late times and moves them closer to the observed values at the NH_3 peak.

Therefore, we conclude that the molecular abundance gradients and the variation of the degree of deuterium fractionation along the TMC-1 ridge can be explained by a chemical evolutionary age difference of more than 10^5 yr or a small increase in the depletion factor of carbon and oxygen, which is itself likely an effect of evolution. Based on the recent mapping observations of DNC and $H^{13}NC$ toward nearby dark cloud cores including the TMC-1 ridge, Hirota et al. (2001) also suggested qualitatively that the systematic variation of the deuterium fractionation could be interpreted in terms of the chemical evolution of dark cloud cores.

It may be interesting to compare the present evolutionary age difference of more than 10^5 yr between the ammonia peak and the cyanopolyyne peak with the results obtained from other models. Howe, Taylor et al. (1996) inferred the age difference between the cores to be of the order of 3×10^5 yr by applying the observed column densities of CCS and its related species to their sequential core collapse model.

Recently, Markwick et al. (2000) proposed another model to explain the observed gradients along the ridge. The protostellar object IRAS 04381+2540 near the ammonia peak produces MHD waves. These Alfvén waves trigger the spontaneous explosion of UV-photolyzed grain mantles by heating the dust grains and their ice mantles through grain–grain collisions. The removal of icy grain mantles restarts the chemical processes in the cores, first in the ammonia core and later in the CP core. The propagation time difference from the *IRAS* source to the cloud cores is 1.5×10^5 yr assuming the speed of MHD waves to be 2 km s^{-1}. This dynamical chemical model produces the gradients in molecular abundances along the TMC-1 ridge. It is interesting to see how deuterium fractionation at both peaks varies with this model, an analysis which has not yet been undertaken. Ambipolar diffusion tends to suppress the contribution of H_2D^+ to gas phase deuterium fractionation (Markwick et al. 2000), although there is an initially high deuterium concentration just after the grain mantle explosion that declines on a timescale of 10^5 yr (Charnley, Tielens, & Millar 1992).

When this paper was in the reviewing process, we learned that Turner (2001) carried out observations of several deuterated species in the dark clouds including TMC-1 and determined precise relative abundances of the deuterated species from an analysis using reliable radiative transport models with microturbulent cloud structure. He reported $R(HCO^+)$ only at the cyanopolyyne peak to be 1.2%, which could be compared with 1.7% ± 0.3% (Caselli et al. 1998). He also studied the chemistry of deuterated species using the modified New Standard Model with several different physical conditions but could not obtain a good match at steady state between the observed and calculated $R(HCO^+)$ for TMC-1CP.

In conclusion, it is shown that the degree of deuterium fractionation of HCO^+ is a plausible candidate to estimate the chemical evolutionary age of dark cloud cores, even though the chemical model used in the present study suffers from some ambiguity in the physical conditions applied, and the errors in the observed relative abundances of DCO^+ are relatively large. Our analysis is based on the fact that the production and destruction of HCO^+ and DCO^+ are well known and relatively simple, and somewhat free from the details of chemical reactions not directly related to deuterium fractionation. The fractionation of DCO^+ thus provides a better constraint on the evolutionary state than other molecular abundances and other deuterated species. If several deuterated species as well as their parent species are observed toward dark cloud cores where physical conditions are well known, we can choose chemical species especially sensitive in their degree of deuterium fractionation to evolutionary age. Furthermore, we can check the reliability of the estimated evolutionary age by using multiple molecular species. Here it must be emphasized that a very reliable chemical model as well as plausible physical conditions are essential in the evolutionary age determination of dark cloud cores.

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