

PHOTOCHEMISTRY AND LIFETIMES OF INTERSTELLAR MOLECULES

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Received 1971 May 10; revised 1971 August 16

ABSTRACT

This paper is concerned with a quantitative discussion of the photochemistry and lifetime against photodecomposition of five interstellar molecules: H_2CO , NH_3 , H_2O , CH_4 , and CO . For the first four molecules, primary photochemical decomposition processes yielding atomic and molecular hydrogen are considered in addition to photoionization. The quantum yield for decomposition of these molecules is unity. For CO , only decomposition to ground-state carbon and oxygen atoms occur, but the quantum yield is not known and may be considerably less than unity. For the interstellar radiation field, we adopt the results of Habing who obtains a uniform energy density $U_\lambda = 40 \times 10^{-18} \text{ ergs cm}^{-3} \text{ \AA}^{-1}$. We estimate the radiation field in obscuring clouds, using an interstellar extinction curve for the Perseus region. The calculations indicate that in unobscured regions all molecules considered here except CO have comparable lifetimes of less than 100 years. The CO lifetime will be 10^2 – 10^3 years for quantum yields of 1–0.1. These short lifetimes suggest that the molecules can never have been exposed to unobscured radiation from the time of their formation until protected in clouds. It implies that polyatomic molecules were formed or released in the gas phase in the clouds where they now occur. Calculations of lifetime as a function of cloud opacity show that for clouds of moderate opacity (a few magnitudes extinction in the visual), polyatomic molecules have lifetimes against photodecomposition of the order of 10^6 years.

I. INTRODUCTION

As of 1971 August the detection of fourteen polyatomic molecules in the interstellar medium has been reported. To the group of molecules detected by radio astronomy we may tentatively add CH_4 , since there are indications that it may have been detected by absorption in the infrared (Herzberg 1968), and several interstellar molecules contain the CH_3 group. A review by Snyder and Buhl (1970) gives the status as of 1970 November. These observations raise questions concerning the mechanism of molecule formation and their subsequent fate in the interstellar radiation field. While the former problem remains obscure, there is little doubt concerning the latter: once formed, the molecules will be subject to destruction by interstellar photons ($\lambda > 912 \text{ \AA}$). This paper is concerned with a quantitative discussion of the photochemistry and lifetimes of stable interstellar molecules.

For an interstellar molecule the lifetime against photodecomposition depends upon three factors: the absorption cross-section, the quantum yield or probability for dissociation following absorption, and the interstellar radiation field. The first two involve the photochemistry of the molecule and are discussed in the following section while the radiation field is discussed in § III. Section IV presents the method of calculation and results for the lifetimes. The final section discusses briefly various aspects of the results.

II. PHOTOCHEMISTRY OF INTERSTELLAR MOLECULES

The absorption cross-section and the photochemical decomposition of NH_3 , H_2O , CO , and CH_4 have been extensively investigated. The references for the absorption cross-sections for each of these molecules is summarized in Table 1. For H_2O , CH_4 , and CO it was necessary to interpolate the data over relatively small wavelength intervals not covered by the references cited. Most of the work on the spectroscopy and photochemistry of H_2CO has been limited to $\lambda > 2000 \text{ \AA}$. Few experiments have been performed below

TABLE 1
REFERENCES FOR ABSORPTION CROSS-SECTIONS σ
OF INTERSTELLAR MOLECULES

Molecule	Wavelength Interval ($\text{\AA}$)	Reference
H ₂ CO	912–1850 2000–3600	Gentieu and Mentall (1970) Calvert and Pitts (1966)
NH ₃	912–1050 1050–2200	Watanabe and Sood (1965) Samson and Myer (1969)
H ₂ O	912–988 1050–1850	Metzger and Cook (1964) Samson and Myer (1969)
CH ₄	912–1000 1150–1600	Metzger and Cook (1964) Samson and Myer (1969)
CO	912–1002 1050–1115	Cook <i>et al.</i> (1965) Myer and Samson (1970)

2000 $\text{\AA}$ where the strong absorption occurs. Indeed, there was not a single quantitative study of the photodecomposition of H₂CO for $\lambda < 2000$ $\text{\AA}$, and the available absorption data (mainly photographic) either were qualitative or covered a very limited wavelength range. The recent determinations in our laboratory of the absorption coefficient for formaldehyde vapor from 650 to 1850 $\text{\AA}$ (Gentieu and Mentall 1970) and a study of the photochemistry of formaldehyde at 1470 and 1236 $\text{\AA}$ (Glicker and Stief 1971) are reported in detail elsewhere. With this information on H₂CO and the published data for NH₃, H₂O, CO, and CH₄, we may now examine the photochemistry of these five molecules as it relates to the interstellar medium. Most of the other observed molecules were discovered after our calculations were made. Although laboratory data are not available for all the remaining molecules, there is no reason to expect their behavior to differ significantly from the polyatomic molecules discussed here.

a) Formaldehyde

The average absorption cross-section for H₂CO is 1000 times greater in the vacuum-ultraviolet region than in the near-ultraviolet, and therefore the photochemistry and lifetime of interstellar formaldehyde will be determined by photon absorption below 2000 $\text{\AA}$. From a study of the photodecomposition of H₂CO at 1470 and 1236 $\text{\AA}$ (Glicker and Stief 1971), evidence was obtained for two primary processes,



both of which occurred with a primary quantum yield (molecules decomposed per photon absorbed) of 0.5. No evidence was obtained for any substantial contribution from the process



which, together with process (1), is important for $\lambda > 2000$ $\text{\AA}$ (Calvert and Pitts 1966). The sum of the quantum yields for decomposition by processes (1) and (2) is unity; i.e., every photon absorbed leads to decomposition of H₂CO. While this may be safely assumed for a molecule having only continuous absorption (i.e., H₂O or CH₄), it need not be so for molecules with a considerable amount of discrete absorption (e.g., CO, H₂CO, and to a lesser extent NH₃).

b) *Ammonia*

The photodecomposition of ammonia proceeds via the following primary processes (McNesby and Okabe 1964; Calvert and Pitts 1966):



whose relative importance depends on the wavelength of light absorbed. Thus process (4) yielding $\text{NH}_2 + \text{H}$ is the sole primary process at 2062 Å (Groth, Schurath, and Schindler 1968; Schurath, Tiedman, and Schindler 1969) and 1849 Å (McNesby, Tanaka, and Okabe 1962) whereas process (5) yielding $\text{NH} + \text{H}_2$ accounts for about 13 percent of the primary photodissociation at 1470 Å (Groth, Okabe, and Rommel 1964) and 1236 Å (McNesby *et al.* 1962), respectively. Presently available data do not allow an estimate to be made of the relative importance of processes (4) and (6) at 1236 Å. Processes (4) and (6) are analogous to processes (3) and (2) in H_2CO photolysis. These results thus suggest that NH_2 and to a lesser extent NH , which cannot form by two-body radiative association, may be present in the interstellar medium as a result of NH_3 photodecomposition.

The only evidence directly relating to the question of the number of NH_3 molecules decomposed per photon absorbed comes from the studies at 2062 Å (Schurath *et al.* 1969). In this study the photolysis of NH_3 was carried out in the presence of an interceptor or scavenger which transformed the primary fragments (H and NH_2) into stable characteristic products. From a quantitative determination of these products, the primary quantum yields of H and NH_2 were estimated to be 0.9 and 0.7–0.9, respectively. Thus the primary quantum yield of decomposition is close to unity.

c) *Water*

The photodecomposition of water may be considered in terms of two primary processes (McNesby and Okabe 1964; Calvert and Pitts 1966):

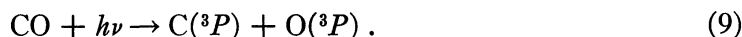


Formation of H and OH is the dominant process at all wavelengths, although formation of H_2 and electronically excited atomic oxygen makes an important contribution (~ 25 percent of the primary process) at 1236 Å (McNesby *et al.* 1962). At 1470 Å, process (7) accounts for from 94 percent (Stief 1966) to 100 percent (Cotton, Masanet, and Vermeil 1966) of the primary process. Thus the principal products of the photodecomposition of water in the interstellar medium are H and OH .

For a molecule such as H_2O with an essentially continuous absorption, all electronic transitions are probably to repulsive states and it may be safely assumed that every photon absorbed leads to dissociation. There is some experimental evidence that this is true for photolysis at 1470 Å (Cotton *et al.* 1966).

d) *Carbon Monoxide*

The bond energy of CO is 11.1 eV, and it is therefore impossible to decompose CO directly with photons of wavelength greater than 1115 Å. Only interstellar radiation in the narrow region 912–1115 Å will be effective. The only spin-allowed primary process in this region is formation of carbon and oxygen atoms in their ground states,



The strength of the absorption for much of this region indicates an allowed transition.

There has been no laboratory work reported on the photolysis of CO below 1115 Å. A quantitative estimate cannot at present be made of the dissociation probability or quantum yield of decomposition. It most certainly will be strongly wavelength dependent. The absorption spectrum of CO exhibits weak bands in the region 1115–1050 Å (Myer and Samson 1970) and strong bands beginning around 1000 Å (Huffman, Larabee, and Tanaka 1964; Cook, Metzger, and Ogawa 1965). An apparent continuous absorption has been observed, but its magnitude and long-wavelength limit are difficult to determine. Photodissociation will occur in the continuum or by predissociation in the banded region. Available data are not sufficient to indicate whether predissociation occurs in the intense bands below 1000 Å. High-resolution spectra of the comparatively weak fourth positive bands in the region above 1060 Å have been reported by Simmons, Bass, and Tilford (1969) not to show any diffuseness or any sharp cutoff of rotational structure. For the entire 912–1115 Å region the dissociation probability or primary quantum yield of CO decomposition could be considerably less than unity.

e) Methane

The photochemistry of methane has been discussed in terms of the primary processes (McNesby and Okabe 1964; Calvert and Pitts 1966):



The dominant process at all wavelengths studied is process (10) yielding molecular hydrogen and the methylene radical (Laufer and McNesby 1968; Gorden and Ausloos 1967). The relative quantum yield of the minor process

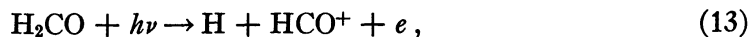


increases about threefold when the wavelength is reduced from 1236 Å to 1048–1067 Å (Gorden and Ausloos 1967).

Methane has a continuous absorption spectrum in the vacuum-ultraviolet, and it may be assumed that every photon absorbed leads to dissociation. This is confirmed by the careful quantum-yield measurement of Laufer and McNesby (1968) which, combined with the data of Gorden and Ausloos (1967) on the relative importance of processes leading to CH, CH₃, and H₂, demonstrate that the sum of the quantum yields for the known primary processes is 0.9 for photolysis at 1236 Å.

f) Photoionization

In addition to the photodecomposition processes discussed above, interstellar molecules may be ionized as a result of interaction with the interstellar radiation field. Table 2 summarizes the available data on the ionization potential of these molecules and gives the wavelength corresponding to the ionization threshold. It is evident that for $\lambda > 912$ Å, the formation of parent molecule ions is energetically possible for all the molecules considered here with the exception of CO and probably HCN. Ionization will in general be somewhat less important than fragmentation since the average ionization efficiencies for NH₃, H₂O, and CH₄ from their ionization thresholds to 912 Å are approximately 40 (Watanabe and Sood 1965), 33, and 10 percent, respectively (Metzger and Cook 1964). Ionization will be even less important in clouds because of attenuation of the short-wavelength radiation. Simultaneous fragmentation and ionization upon single-photon absorption is not energetically possible at $\lambda > 912$ Å with the possible exception of the process



which has a threshold at 13.4 eV (925 Å).

TABLE 2
IONIZATION POTENTIALS AND THRESHOLD WAVELENGTHS FOR
IONIZATION OF INTERSTELLAR MOLECULES

Molecule	Ionization Potential (eV)	λ (Å)	Reference
NH ₃	10.2	1215	Watanabe (1957)
CH	10.6	1170	Herzberg and Johns (1969)
H ₂ CO	10.9	1137	Watanabe (1957)
CH ₃ OH	10.9	1137	Watanabe (1957)
HCOOH	11.1	1117	Watanabe (1957)
H-C≡C-CN	11.6	1069	Diebler, Reese, and Franklin (1961)
H ₂ O	12.6	984	Watanabe (1957)
CH ₄	13.0	954	Watanabe (1957)
OH	13.1	946	Foner and Hudson (1958)
HCN	13.6	912	Diebler and Liston (1968)
H	13.6	912	
CO	14.0	886	Watanabe (1957)
CN	14.2	873	Diebler and Liston (1967)

III. THE INTERSTELLAR RADIATION FIELD

The interstellar radiation field which a molecule sees will vary considerably depending primarily upon the degree of obscuration in the vicinity of the molecule. As we are concerned with continuous rather than line absorption, only the grains will contribute. The effect of interstellar extinction and its spatial fluctuations on interstellar chemistry must be examined.

In the so-called clear regions of the Milky Way the extinction in the visible is a few tenths of a magnitude per kiloparsec (Bok 1936). We refer to these regions as unobscured. Several calculations of the interstellar radiation field appropriate to the unobscured regions have been carried out. These are in general agreement except near $L\alpha$ for which Lambrecht and Zimmermann (1956) have a peak several times greater than the rather flat distribution of other investigators. Their result, which approximately doubles the ultraviolet flux, does not significantly increase the uncertainty of the calculation. We adopt here the results of Habing (1968), who obtains an ultraviolet energy density U_λ between 30 and 50×10^{-18} ergs cm⁻³ Å⁻¹ between 1000 and 2200 Å. Lambrecht and Zimmermann's (1956) results for wavelengths to 3646 Å are sufficiently consistent with this value that we have adopted a constant $U_\lambda = 40 \times 10^{-18}$ ergs cm⁻³ Å⁻¹ for $\lambda > 912$ Å. This calculated density agrees very well with the measured flux of Lillie (1968) for the same wavelength range. The uncertainty of the calculations and the small effect on the lifetimes do not warrant the additional work of allowing for any wavelength dependence.

Radio observations of interstellar molecules permit their spatial distribution to be determined and indicate an association with obscured regions. Consequently, it is necessary to determine the radiation field in obscuring clouds. This has been done by combining the adopted constant flux with an interstellar extinction curve covering the visible and ultraviolet regions. For the visible we used Johnson's (1965) curve for the Perseus region; for the ultraviolet, Stecher's (1969) curve determined from rocket observations of ζ and ϵ Per. These results are in general agreement with the satellite observations of Code (1969). In both wavelength intervals, regional variations in the extinction curve are found. Errors are introduced by using data from only one region for the entire Milky Way. However, the Perseus curve appears to be applicable within an ac-

ceptable uncertainty to several other areas. An uncertainty difficult to evaluate arises from the assumption that grains in the low-density, relatively unobscured regions have the same optical properties as in much more highly obscured, denser clouds where the molecules are found. Although these factors in the analysis should be kept in mind, we will go ahead on the hypothesis that the final results are qualitatively correct and that the uncertainty is not large enough to affect conclusions regarding the stability and origin of molecules. It will be seen from the results that this hypothesis is justified.

Stecher's and Johnson's curves overlap at the U point. From Table 5 of Johnson, $A_U = 4.7$ for $B - V = 1$, to which value the ultraviolet curve is normalized. Figure 1 presents the Perseus extinction curve on an absolute basis. This was obtained by re-drawing Stecher's curve with the ordinate displaced to make $A_U = 4.7$, extrapolating the curve to $1/\lambda = 10 \mu^{-1}$ and using Johnson's curve for the long-wavelength end.

We next determined the radiation field which has penetrated zones of increasing optical thickness measured by the visible extinction A_v . Steps of 1.0 in A_v were chosen and the extinction determined at a series of wavelengths to 900 Å by multiplying the normalized set by $\frac{1}{3}A_v$ ($A_v = 3$ on the normalized curve of Fig. 1). These magnitudes were converted into transmissivities. Figure 2 is a plot of transmissivity as a function of wavelength for various optical depths in a cloud. These transmissivities may be multiplied by the adopted uniform interstellar radiation field to obtain the radiation field as a function of cloud depth.

IV. LIFETIME OF INTERSTELLAR MOLECULES

The probability P that a molecule is decomposed by light in the interstellar medium is given by

$$P = \int N_\lambda \Phi_\lambda d\lambda, \quad (14)$$

where N_λ = number of photons absorbed by the molecule in the interval $d\lambda$ and the limits of integration are from 912 Å to the photodissociation threshold, and Φ_λ = the primary quantum yield of decomposition (i.e., the probability of dissociation per absorbed photon).

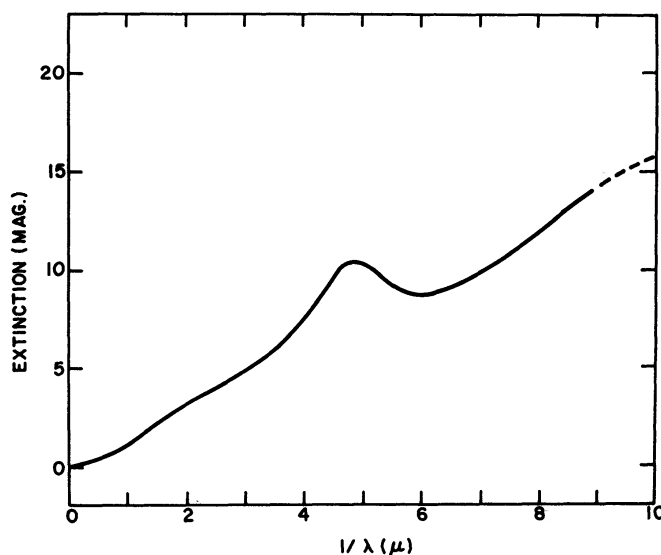


FIG. 1.—Extinction curve for the Perseus region. Dotted portion is an extrapolation

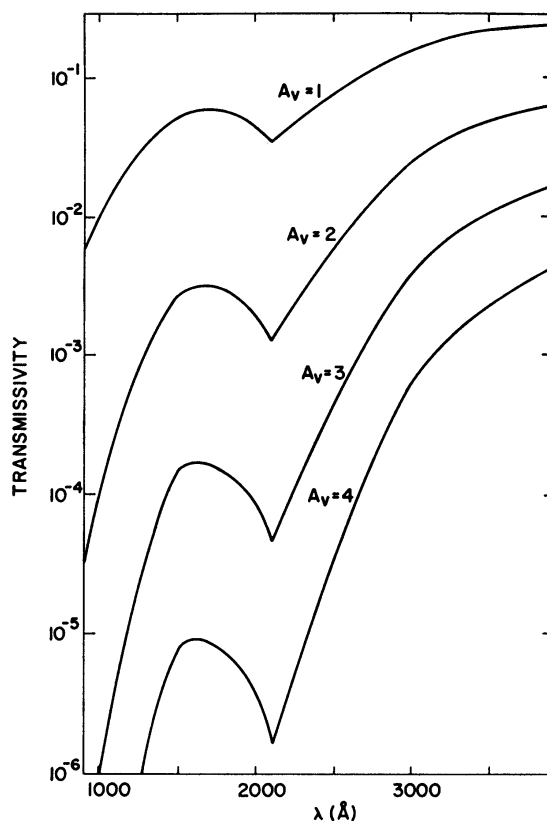


FIG. 2.—Transmissivity in cloud as a function of wavelength. A_V is the cloud extinction in magnitudes at 5500 Å.

In terms of the energy density U_λ , for an isotropic radiation field equation (14) becomes

$$P = h^{-1} \int U_\lambda \sigma_\lambda \Phi_\lambda d\lambda, \quad (15)$$

where h = Planck's constant. The lifetime of the molecule against photodecomposition is simply the reciprocal of the dissociation probability P .

P is determined by integration of the product over all wavelengths from 912 Å to the photodissociation threshold of the molecule. However, as discussed in the last section, it is likely that U_λ does not vary by more than a factor of about 2 over the wavelength interval 1000–3600 Å and, with the exception of cloud areas, also does not vary by more than a small factor throughout interstellar space. Therefore, in interstellar space free of obscuring clouds the dissociation probability can be written

$$P = \frac{U\Phi}{h} \int \sigma_\lambda d\lambda, \quad (16)$$

where U is the integrated energy density.

For all molecules discussed in § II except carbon monoxide we put $\Phi = 1$. Carbon monoxide lifetimes were calculated by using both $\Phi = 1$ and $\Phi = 0.1$.

It is obvious that lifetimes will be orders of magnitude longer in clouds where there is high obscuration of interstellar radiation. In order to examine the effect of increasing

depth of the cloud, we have calculated the lifetimes of H_2CO , H_2O , NH_3 , CO , and CH_4 as a function of increasing optical thickness. The latter is measured in terms of the extinction in the visual, A_v , as discussed in § III and shown in Figure 2 as a plot of transmissivity T_λ versus λ for steps of 1.0 in A_v .

The dissociation probability for an obscured region is given by

$$P = \frac{U\Phi}{h} \int T_\lambda \sigma_\lambda d\lambda = U \langle T\sigma \rangle \Phi / h. \quad (17)$$

The procedure was to plot the product $T\sigma\lambda$ versus λ and obtain $\langle T\sigma \rangle$ from the area under the curve. In practice, both of these were done numerically on a computer with areas determined by the spline interpolation method (Thompson 1970; Scudder 1970). Comparison with values determined by hand plotting and area measurements with a planimeter showed agreement to within 10 percent for lifetimes in unobscured regions.

Figure 3 shows lifetimes against photodecomposition in unobscured regions (represented by $A_v = 0$) and in regions with increasing degrees of obscuration ($A_v = 1, 2, 3$, and 4 mag).

V. DISCUSSION

The results in Figure 3 indicate that in clear interstellar regions all molecules except carbon monoxide have comparable lifetimes of less than 100 years. These are all extremely short on the galactic time scale. Thus, the molecules water, ammonia, methane, and formaldehyde can exist only in dense clouds which protect them from the full interstellar radiation field. This is consistent with observations of ammonia (Cheung *et al.* 1969) and formaldehyde (Zuckerman *et al.* 1970). Further, these molecules can never

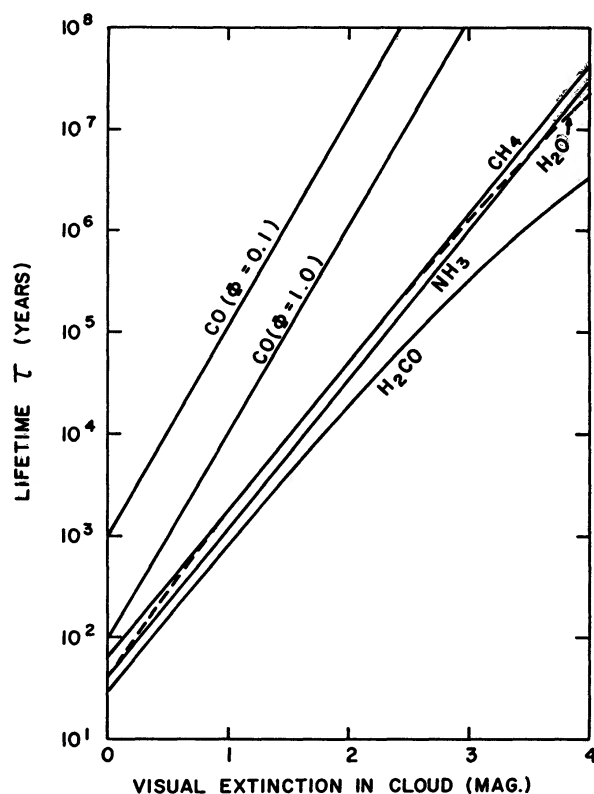


FIG. 3.—Lifetimes of interstellar molecules in clouds

have been exposed to the unobscured radiation from the time of formation until protected in clouds. This requirement imposes a severe restriction on possible mechanisms of formation. It implies that polyatomic molecules were formed or released in the gas phase in the clouds where they now occur.

The lifetime obtained here for NH_3 ($\tau = 40$ years) is less than that estimated by Cheung *et al.* (1968) ($\tau = 100$ years). The difference is due to their use of a cross-section ($3.7 \times 10^{-18} \text{ cm}^2$) given by Potter and Del Duca (1964) for the region down to $1216 \text{ \AA}$. The interstellar radiation field extends down to $912 \text{ \AA}$, and the average absorption cross-section for NH_3 in the region $912\text{--}1050 \text{ \AA}$ is 3 times larger than it is in the region $1050\text{--}2200 \text{ \AA}$. This results in a larger dissociation probability P and hence a shorter lifetime.

The carbon monoxide lifetime is 2–20 times longer than that for the other molecules considered here, depending upon the value of the quantum yield. The CO molecules therefore require considerably less protection than formaldehyde and the other interstellar molecules. This property can readily explain the observation (Wilson, Jefferts, and Penzias 1970) that the size of the carbon monoxide cloud is at least an order of magnitude larger than that of a typical formaldehyde cloud. The smaller quantum yield for carbon monoxide leads to a lifetime of 10^3 years, which is comparable to that of molecular hydrogen (Stecher and Williams 1967) and the OH and CH radicals (Hesser and Lutz 1970).

Stecher and Williams (1966) give a rough estimate of 10^6 years for the lifetime of CO in a $10,000^\circ \text{ K}$ radiation field with a dilution factor of 3×10^{-15} . We have corrected this to the uniform Habing field and obtain 10^4 years. For this to be in agreement with the results of Figure 3, the quantum yield Φ would have to be 10^{-2} .

Figure 3 shows, as expected, that the lifetimes of molecules in clouds with a few magnitudes extinction become the order of 10^6 years. Contrary to the lifetime of less than 100 estimated in the cloud-free regions, lifetimes in clouds of moderate opacity are sufficiently long for the molecules to be more than transient residents of interstellar space. It is also worth noting that the order of lifetimes in the denser clouds is $\text{CH}_4 > \text{H}_2\text{O} \cong \text{NH}_3 > \text{H}_2\text{CO}$. This is expected, since the long-wavelength cutoff of absorption increases in this same order—viz., 1600, 1850, 2200, and $3800 \text{ \AA}$, respectively, for CH_4 , H_2O , NH_3 , and H_2CO —and the degree of protection is reduced.

In addition to photodissociation and any photoionization that occurs, molecules can also be destroyed by interaction with high-energy radiation (X-rays and γ -rays) and energetic particles. These processes are much less effective than destruction by ultraviolet radiation. However, when the ultraviolet is highly attenuated in clouds, the energetic radiation and particles will persist and become relatively more important. The ultimate lifetime in clouds may depend upon these processes and may be shorter than those shown in Figure 3. These processes require further study.

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