PHOTOCHEMISTRY OF INTERSTELLAR MOLECULES

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PHOTOCHEMISTRY OF INTERSTELLAR MOLECULES

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Abstract-

Photodissociation is the primary destructive process for interstellar molecules outside of dense clouds. A quantitative discussion of the photochemistry of two diatomic and eight polyatomic molecules is given. Six are known to occur in the interstellar medium and the others are suspected or otherwise of interest. For an interstellar molecule the lifetime against photodecomposition depends upon three factors: the absorption cross section, the quantum yield or probability of dissociation following photon absorption, and the interstellar radiation field. The first two factors plus the mode of photodecomposition or photochemical primary processes constitute the photochemistry of the molecule. For some of these molecules the pathways for decomposition or the quantum yield is uncertain and each may depend upon wavelength. In the case of CO, the only spin allowed process energetically possible with interstellar photons (\( \lambda > 912 \ \text{\AA} \)) is formation of carbon and oxygen atoms in their \( ^3 \text{P} \) ground states. No laboratory studies of CO photolysis have been reported for wavelengths below the decomposition threshold at 1115 \( \text{\AA} \). If long-lived excited states are involved below 1115 \( \text{\AA} \) as has been shown to be at longer wavelengths, the dissociation probability could be significantly less than unity.

The third factor influencing the lifetime of an interstellar molecule is the interstellar radiation field. For the unobserved regions we adopt the results of Habing who obtains a constant energy density of \( 4 \times 10^{-17} \text{ ergs} \).
The radiation field in obscuring clouds is estimated by combining the adopted constant flux with the observed interstellar extinction curve covering the visible and ultraviolet regions.

Lifetimes against photodecomposition in the unobscured regions and as a function of increasing optical thickness in obscuring clouds are calculated for the ten species. The results show that, except for CO, all the molecules considered here have comparable lifetimes of less than one hundred years in unobscured regions. Thus these molecules can exist only in dense clouds and can never have been exposed to the unobscured radiation. This implies that polyatomic molecules were formed or released in the gas phase in the clouds where they now occur. The calculation further show that the lifetimes in clouds of moderate opacity (a few magnitudes extinction in the visual) are of the order of $10^6$ years. The lifetime for CO in unobscured regions will be between one hundred and one thousand years if the dissociation probability is between unity and one-tenth. The smaller dissociation probability leads to a lifetime comparable to that for other diatomics. This means that CO molecules require considerably less protection than formaldehyde and the other polyatomics and readily explains the observation that the size of a CO cloud is at least an order of magnitude larger than that of a typical formaldehyde cloud.
I. INTRODUCTION

Within the last three years five diatomic and some fourteen polyatomic molecules have been discovered in interstellar space, mainly by radio astronomy. Prior to this, only a few diatomic species were observed, mainly by optical astronomy in the visible region. This unexpected increase in both the number and complexity of interstellar molecules is evidence of significant chemical phenomena associated with the interstellar medium. While the interaction of many scientific disciplines is required e.g. astronomy, astrophysics, spectroscopy, photochemistry, chemical kinetics, surface chemistry, etc., I wish to discuss here the role of photochemistry in determining the lifetime of interstellar molecules.

The photochemistry and lifetimes have previously been discussed (Stief, Donn, Glicker, Gentieu and Mentall, 1972) for the molecules $\text{H}_2\text{CO}$, $\text{NH}_3$, $\text{H}_2\text{O}$, $\text{CH}_4$ and CO. This has been extended to include new molecules detected since the first calculations were made ($\text{OCS}$, $\text{CH}_3\text{C} \equiv \text{CH}$), one molecule (NO) which has been the subject of an extensive negative search, and two other molecules ($\text{C}_2\text{H}_2$ and $\text{C}_6\text{H}_6$) which might be expected to differ from the usual pattern.

For an interstellar molecule the lifetime against photodecomposition depends upon three factors: the absorption cross section, the quantum yield or probability of dissociation following photon absorption, and the interstellar radiation field. The first two factors plus the mode of photodecomposition or primary photochemical process constitute the photochemistry of the molecule and this will be discussed in the next section. The radiation field adopted will be discussed briefly in Section III while Section IV will present the calculations on lifetime against photodecomposition. Finally, some discussion will be given of the implications of these results.
II. Photochemistry of Interstellar Molecules

The quantitative spectroscopy and photochemistry of the ten molecules considered here have been investigated in the laboratory to varying degrees. References for the absorption cross sections used in the lifetime calculation are collected in table I. The photochemistry of the molecules \( \text{H}_2\text{CO}, \text{NH}_3, \text{H}_2\text{O}, \text{CH}_4 \) and CO as it applies to the interstellar medium has been discussed previously (Stief, et al, 1972) and will only be summarized here. The important primary processes for the first four molecules are formation of atomic hydrogen and formation of molecular hydrogen; the relative importance of these decomposition channels is usually strongly dependent on wavelength. Presently available evidence indicates that the quantum yield of dissociation or decomposition probability is unity for \( \text{H}_2\text{CO}, \text{NH}_3, \text{H}_2\text{O} \) and \( \text{CH}_4 \).

Carbon monoxide is unique in that no laboratory studies of its photo-decomposition have been reported for wavelengths below the decomposition threshold at \( 1115\AA \). The only spin allowed process energetically possible with interstellar photons (\( \lambda > 912 \AA \)) is formation of carbon and oxygen atoms in their \( \text{^3P} \) ground states. If long-lived excited states are involved below \( 1115\AA \) as have been shown to be at longer wavelengths, the dissociation probability could be significantly less than unity.

The photochemistry of the remaining five molecules will now be considered separately in more detail.

A. Carbonyl Sulfide

The photochemical decomposition of OCS may be considered in terms of the primary processes:
OCS + hv → CO + S \hspace{1cm} (1)

→ CS + O \hspace{1cm} (2)

From the studies of Sidhu, Csizmadia, Strausz and Gunning (1966) at 2537 and 2288Å, the quantum yield for process (1) is 0.90 with formation of S(1D) predominant over S(3p). The preferential formation of S(1D) would be expected on the basis of spin conservation.

Formation of atomic oxygen via processes (2) becomes energetically possible at 1753 Å for O(3p) and 1360 Å for O(1D). Calvert and Pitts (1966) cite qualitative evidence for the occurrence of (2) at very short wavelengths. The process OCS → C + O + S is not energetically possible for interstellar photons (λ > 912Å). The ionization potential of OCS is 11.2 eV (Matsunaga and Watanabe, 1967) which corresponds to an ionization threshold of 1110 Å. The ionization efficiency is about 50% in the region below 1100 Å.

B. Methyl Acetylene

Photolysis in the near ultraviolet continuum (2000-1600Å) occurs via the primary process

\[ \text{CH}_3\text{C} \equiv \text{CH} + \text{hv} \rightarrow \text{CH}_2\text{C} \equiv \text{CH} + \text{H} \hspace{1cm} (3) \]

Ramsay and Thistlethwaite (1966) have observed the propargyl radical (CH₂C ≡ CH) in absorption during flash photolysis of methyl acetylene and the products observed by Galli, Harteck and Reeves (1967) at 2062Å are consistent with process (3). No quantitative data on the quantum yield of decomposition is available for absorption in the continuum.

In the banded region below 1600 Å, Stief, De Carlo and Payne (1971)
found evidence at 1470Å for processes leading to atomic hydrogen ($\Phi \geq 0.40$) and molecular hydrogen ($\Phi = 0.15$). Although several processes probably occurred, it is important to note here that the primary quantum yield of dissociation must be 0.65 or larger. At 1236Å, Payne and Stief (1971) found that molecular hydrogen formation occurred to the exclusion of atomic hydrogen formation. An important process was found to be

$$\text{CH}_3\text{C} \equiv \text{CH} + \hbar \nu \rightarrow \text{C}_3^+ + 2\text{H}_2$$

which occurs via the short-lived $\text{C}_3\text{H}_2$ radical.

Interstellar photons ($\lambda > 912\text{Å}$) are capable of ionizing methylacetylene since the ionization potential of 10.36 eV (Nakayana and Watanabe, 1964) corresponds to $\lambda = 1196\text{Å}$. The ionization efficiency may be estimated as 50 - 75% below 1175 Å.

C. Nitric Oxide

The photodissociation of nitric oxide has been discussed (Calvert and Pitts, 1966) in terms of the following primary processes:

$$\text{NO} + \hbar \nu \rightarrow \text{N} + \text{O}$$
$$\rightarrow \text{NO}^+ + \text{e}$$

McNesby and Okabe (1964) discuss the evidence that predissociation begins above $v' = 7$ of the $B^2\Pi$ state, corresponding to 6.5 eV or 1910 Å. Hence, only interstellar photons below 1910Å are capable of dissociating NO. Ionization occurs below 1338Å based on Dressler and Miescher's (1965) value.
for the ionization potential. The data of Watanabe, Matsunaga and Sakai (1967) indicate that the ionization efficiencies are mostly in the range 40 - 80% for $\lambda = 912$ to 1300 Å.

There is little direct information available on the primary quantum yield. From the low pressure data of Leiga and Taylor (1965), the product quantum yields for photolysis of NO at 1236 Å, 1470 Å and the region 1550-1650 Å are at least not inconsistent with primary quantum yields in the range 0.7 to 0.9.

D. Acetylene

The photochemistry of acetylene may be considered in terms of the following primary processes:

$$C_2H_2 + h\nu \rightarrow C_2H + H$$  \hspace{1cm} (7)

$$\rightarrow C_2 + H_2$$  \hspace{1cm} (8)

$$\rightarrow C_2H_2^*$$  \hspace{1cm} (9)

The data of Zelikoff and Aschenbrand (1956) at 1849 Å lead to a value of approximately 0.2 for the quantum yield of process 7. Stief, De Carlo and Mataloni (1965) suggest that the quantum yield of process 8 is of the order of 0.1 at both 1470 and 1236 Å. In the laboratory, the long-lived excited acetylene molecule formed in process 9 reacts with excess acetylene to form a variety of products (Takita, Mori and Tanaka, 1969); this has little application to the photodecomposition of interstellar molecules. The observation at low resolution of a broad, structureless emission in the region 4000 to 6000 Å during photolysis of C$_2$H$_2$ at 1236 Å was tentatively identified by Stief, De Carlo and Mataloni (1965) with
the Swann bands of $C_2$. The recent observation by Becker, Haaks and Schurgers (1971) of this same broad emission feature under high resolution in both the 1236 Å photolysis of $C_2H_2$ and in the acetylene-oxygen atomic flame rules out a diatomic molecule as the emitter. Arguments are presented that $C_2H_2$ may be the emitting species. If this is correct, then at low pressures process (9) followed by emission from some low-lying excited state does not lead to dissociation of $C_2H_2$ and the total primary quantum yield may not exceed 0.2 at all wavelengths studied.

The ionization potential of $C_2H_2$ is 11.4 eV (Nakayama and Watanabe, 1964; Diebler and Reese, 1964) and thus photons of $\lambda < 1087\text{Å}$ are capable of ionizing acetylene. Below 1000 Å, the ionization efficiency is approximately 75% (Metzger and Cook, 1964).

E. Benzene

Benzene does not undergo any significant photochemical reactions in the first absorption band (2800 to 2100Å), although important photophysical processes do occur. We therefore restrict our attention to wavelengths below 2100Å. Although several recent photochemical studies have been performed in this spectral region, the complexity of the system is such that detailed knowledge of primary processes is still lacking. Shindo and Lipsky (1966) and Foote, Mallon and Pitts (1966) have shown that the primary quantum yield of dissociation at 1849Å extrapolates at low pressure (0.1 torr) to approximately unity. The quantitative experiments of Hentz and Rzad (1967) at 1 torr pressure show that a minimum value for $\phi$ is 0.30 and 0.15 at 1470 and 1236 Å respectively. Photoionization
occurred at the latter wavelength since the ionization potential of benzene is 9.24 eV (Watanabe, Nakayama and Mottl, 1962) corresponding to an ionization threshold of 1341 Å. Below about 1250 Å, the average ionization efficiency is 50% (Person, 1965).

III. THE INTERSTELLAR RADIATION FIELD

The component of the interstellar radiation field we will be concerned with is the total dilute starlight coming from all stars. Short wavelength radiation from hot stars can ionize the atomic hydrogen in their immediate vicinity. We are not concerned here with these relatively small regions dominated by ionized H but with the larger remaining HI regions where hydrogen is neutral. A more thorough discussion of the interstellar radiation field adopted for these calculations has been given previously (Stief et al, 1972) and will only be summarized here. For unobscured regions we adopt the results of Habing (1968) who obtains an ultraviolet energy density \( U \) between 30 and 50 \( \times 10^{-18} \) ergs \( \text{cm}^{-3} \text{Å}^{-1} \) for wavelengths between 1000 and 2200 Å. Lambrecht and Zimmermann's (1955) results for wavelength to 3646 Å are sufficiently consistent with this value that we have adopted a constant \( U = 40 \times 10^{-18} \) ergs \( \text{cm}^{-3} \text{Å}^{-1} \) for \( \lambda > 912 \) Å. The uncertainty of the calculations and the small effect on the lifetimes do not warrant the allowing for any wavelength dependence.

To determine the radiation field in obscuring clouds, we combine 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 and for the ultraviolet, Stecher's (1969) curve determined from rocket observations of Persei. Figure 1 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 interstellar space free of obscuring clouds is

$$P = \frac{U\phi}{h} \int \sigma_\lambda d\lambda = \frac{U\phi}{h} < \sigma \lambda >$$

where $h$ = Planck's constant, $U$ is the energy density, $\phi$ is the primary quantum yield of dissociation, $\sigma_\lambda$ is the absorption cross section, and the limits of integration are from $912 \, \AA$ to the photodissociation threshold. From the list of interstellar molecules observed to date, we have chosen six for which sufficient data is available: $\text{NH}_3$, $\text{H}_2\text{O}$, $\text{H}_2\text{CO}$, CO, COS and $\text{CH}_3\text{C} = \text{CH}$. Calculations have also been made for four additional molecules which are of interest for a variety of reasons: $\text{CH}_4$, NO, $\text{C}_2\text{H}_2$ and $\text{C}_6\text{H}_6$. Methane is of interest since there are indications it may have been detected by absorption in the infrared (Herzberg, 1968). Nitric oxide has been extensively searched for in some 67 sources (Turner, Heiles and Scharlemann, 1970) with only negative results to date. It has been suggested that this may be due to an unusually short photodissociation lifetime for NO. Acetylene and benzene may have long lifetimes if processes other than bond rupture occur (e.g. internal conversion, fluorescence, phosphorescence, etc). Benzene is also of interest as the simplest aromatic molecule since all the organic molecules observed so far have been aliphatic.

The references for the absorption cross sections for the ten molecules under consideration are summarized in table I. For all the
molecules except $\text{NH}_3$, $\text{NO}$ and $\text{C}_6\text{H}_6$ it was necessary to interpolate the data over relatively small wavelength intervals not covered by the data available; frequently this was in the region 1000 to 1050 Å. For most of the molecules considered, we have taken the quantum yield of dissociation to be unity. This is to be expected for those molecules with essentially continuous absorption and is consistent with available laboratory data. The exceptions are $\text{CO}, \text{C}_2\text{H}_2$ and $\text{C}_6\text{H}_6$. At this stage we will carry $\Phi$ as an unevaluated factor for these three molecules.

From equation 10 and using the laboratory data for $\sigma$, the procedure was to plot $\sigma \lambda$ vs $\lambda$ and obtain $<\sigma \lambda>$ from the area under the curve. Both of these were done numerically on a computer with areas determined by the spline interpolation method (Scudder, 1971; Thompson, 1970).

Table II gives the lifetimes in unobscured regions for the molecules in question.

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 the ten molecules discussed above as a function of increasing optical thickness. The latter is measured in terms of the extinction in the visual, $A_v$, as shown in Figure 1 as a plot of transmissivity $T_\lambda$ vs $\lambda$ for steps of 1.0 in $A_v$. The dissociation probability for an obscured region is given by

$$P = \frac{U\Phi}{h} \int T_\lambda \sigma_\lambda \lambda d_\lambda = \frac{U\Phi}{h} < T\sigma_\lambda >$$  \hspace{1cm} (11)

Similar to the previous calculation, the value of $< T\sigma_\lambda >$ was obtained
via plotting and numerical integration using a computer. Figures 2 and 3 show lifetimes against photodecomposition with increasing obscuration, the latter measured in terms of extinction in the visual ($A_v$).

V. IMPLICATIONS OF THE LIFETIME CALCULATIONS

The results in Table II suggest that in clear interstellar regions, most of the molecules under consideration have lifetimes against photodissociation of the order of 100 years or less. That these lifetimes are extremely short on the galactic time scale is evident from the following. If one assigns to the molecules velocities comparable to those of the clouds as a whole, that is velocities of the order of 10 to 100 km/sec, the total distance traveled by a molecule in its lifetime of 100 years ($3 \times 10^9$ sec) is only $3 \times 10^{15}$ to $3 \times 10^{16}$ cm or 0.001 to 0.01 pc. Since cloud diameters are typically of the order 0.1 to 10 pc and intercloud distances even larger, it is evident that the average polyatomic molecule cannot travel a significant distance in unobscured regions without being subject to destruction by interstellar photons. Thus polyatomic molecules can exist only in dense clouds which protect them from the full interstellar radiation field. This is consistent with observations. Further, these molecules can never 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 cloud where they now occur.
The carbon monoxide lifetime will be $10^2$ to $10^3$ years if the quantum yield $\phi$ for decomposition is 1 to 0.1. An alternate estimate for CO may be made by considering the dissociation continua only, for which $\phi = 1$, and ignoring the possible contribution from predissociation in the banded region. From the continua estimated by Cook, Metzger and Ogawa (1965) from their own data, we calculate $\tau=10^3$ years. This may be longer than the true value if predissociation contributes significantly or shorter if the apparent dissociation continua has been overestimated due to insufficient resolution of the closely spaced bands in the spectrum. Nevertheless, it is evident that carbon monoxide requires 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 and the estimate from the dissociation continua both lead to a lifetime comparable to that for other diatomics.

The benzene and acetylene lifetimes are at least 5 and 20 years respectively if the quantum yields are unity. While it is likely that $\phi$ is in fact somewhat less than unity for both these molecules, it is doubtful if $\phi$ will be sufficiently small to increase $\tau$ much above the typical value of 50 to 100 years. The lifetime for COS appears to be somewhat shorter than the typical value, although it is not certain if the difference is significant.
Figures 2 and 3 show, 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 one hundred years estimated in the cloud-free regions, lifetimes in clouds of moderate opacity are significant compared to the time scale of the clouds themselves.

In addition to photodissociation, the lifetimes calculated here may include a contribution from photoionization. From the ionization potentials of the ten molecules being considered, it is concluded that interstellar photons ($\lambda > 912 \, \text{Å}$) are capable of producing the parent ion for all molecules except CO. Ionization will in general be less important than bond rupture since it occurs over a narrower wavelength range and since ionization efficiencies are less than unity for polyatomic molecules. Ionization will be even less important in clouds due to attenuation of the short wavelength radiation.

In addition to photodissociation and any photoionization that occurs, molecules can also be destroyed by interaction with high energy radiation (X 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 lifetimes in clouds may depend upon these processes and may be shorter than those shown in Figures 2 and 3. These processes require further study.
ACKNOWLEDGMENTS

The author is indebted to Dr. Bertram Donn for his patient tutoring in astronomical problems, especially those concerning the interstellar medium. The present paper owes much to stimulating discussion with him. Thanks are also due to Mr. Thorton Pinder for his help with compiling the data on absorption cross sections and to Mr. William Mish for his expert guidance and assistance in the computer calculations.
REFERENCES

Herzberg, G. 1968, Conf. on Lab. Astrophysics, Lunteren, Neth.


<table>
<thead>
<tr>
<th>Molecule</th>
<th>Reference</th>
</tr>
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<tr>
<td>OCS</td>
<td>Cook and Ogawa (1969); Matsunaga and Watanabe (1967); Sidhu et al (1966).</td>
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<td>$\text{H}_2\text{CO}$</td>
<td>Gentieu and Mentall (1970); Calvert and Pitts (1966).</td>
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<td>$\text{CH}_3\text{C} = \text{CH}$</td>
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<td>Watanabe and Sood (1965); Samson and Myer (1969).</td>
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<td>Metzger and Cook (1964); Samson and Myer (1969).</td>
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<td>$\text{NO}$</td>
<td>Watanabe, Matsunaga and Sakai (1967); Samson and Myer (1969).</td>
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<td>$\text{C}_6\text{H}_6$</td>
<td>Bunch, Cook, Ogawa and Ehler (1958); Marmo (1968).</td>
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<tr>
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<td>Metzger and Cook (1964); Nakayama and Watanabe (1964).</td>
</tr>
<tr>
<td>CO</td>
<td>Cook, Metzger and Ogawa (1965); Myer and Samson (1970).</td>
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Table II. Lifetimes of Interstellar Molecules in Unobscured Regions.

<table>
<thead>
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<th>Molecule</th>
<th>τ(years)</th>
<th>Molecule</th>
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<td>COS</td>
<td>10</td>
<td>H$_2$O</td>
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<td>30</td>
<td>NO</td>
<td>100</td>
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<td>5/§</td>
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<tr>
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<td>40</td>
<td>C$_2$H$_2$</td>
<td>20/§</td>
</tr>
<tr>
<td>CH$_4$</td>
<td>40</td>
<td>CO</td>
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§ = primary quantum yield of dissociation.
CAPTIONS FOR FIGURES

Fig. 1  Transmissivity in cloud as a function of wavelength, $A_v$ in the cloud extinction in magnitudes at 5500 Å.

Fig. 2  Lifetimes of molecules in clouds.

Fig. 3  Lifetimes of molecules in clouds.
LIFETIME $\tau$ (YEARS)

EXTINCTION IN THE VISUAL ($A_v$)

$\text{CO (} \Phi = 0.1 \text{)}$

$\text{H}_2\text{O}$

$\text{H}_2\text{CO}$

$\text{OCS}$

$\text{C}_2\text{H}_2$
EXTINCTION IN THE VISUAL (Av)

LIFETIME $\tau$ (YEARS)

$10^1$ $10^2$ $10^3$ $10^4$ $10^5$ $10^6$ $10^7$ $10^8$

4 3 2 1

CH$_4$ NO NH$_3$

C$_6$H$_6$ C$_3$H$_4$

EXTINCTION IN THE VISUAL (Av)

FIG 3