Photodestruction Rates for Cometary Parent Molecules

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I present new evaluations of the photodestruction rates for several molecules of cometary interest, together with a critical comparison with other estimations, and a summary of the need for future laboratory measurements.

INTRODUCTION

The lifetime of cometary molecules versus photodestruction is a basic parameter for all cometary studies, since it governs the spatial distribution of theses species, and is a requisite for deriving molecular abundances from spectroscopic observations. It is also needed for chemical modeling of planetary atmospheres.

Systematic studies for molecules in interstellar clouds are available [*Lee,* 1984; *van Dishoeck,* 1988; *Roberge et al.,* 1991]. Comprehensive studies for molecules in cometary conditions were made by *Jackson* [1976], *Huebner and Carpenter* [1979], *Huebner* [1985], and *Huebner et al.* [1992]. In several cases, the published rates differed by more than 1 order of magnitude. For several molecules investigated during recent infrared or radio observations, no rates were available. Some analyses of cometary observations were based upon erroneous rates. For many molecular species, improved laboratory data are now available. Thus I feel it would be appropriate to undertake new evaluations of photodestruction rates for cometary parent molecules. When I undertook this study, the monumental work of *Huebner et al.* [1992] was not yet available in its final form. I think that the present paper complements rather than duplicates that work, because it includes several additional species and it updates several rates on the basis of more recent or more complete laboratory measurements.

The new rates evaluated here are for a heliocentric distance of 1 AU, assuming the quiet Sun reference spectrum of *Huebner and Carpenter* [1979]. (The rates are expected to be higher in periods of solar activity. The solar Lyman α flux may then be enhanced by a factor of 2 or even more.) The present review is limited to a selection of potential cometary parent molecules, i.e. to stable volatile molecules liable to be contained in cometary nuclei. The review is presented in the form of a table (Table 1) and of molecule-by-molecule comments. The rates relevant to the present work are listed under the column head "Meudon."

COMMENTS ON INDIVIDUAL MOLECULES

Water Group

H_2O water. The photolysis of water for cometary conditions was reviewed by *Crovisier* [1989] and *Wu and Chen* [1993]. This topic is well documented and well understood, both experimentally and theoretically. The branching ratio of the channel yielding O(^1D), however, is very poorly known. This is unfortunate, since prompt emission of O(^1D) is an important cometary emission which could yield the water production rate.

HDO deuterated water. The absorption spectra of H_2O and HOD are almost the same within the experimental uncertainties. The problem is to determine the branching ratio between HDO + $h\nu \rightarrow$ OH + D and OD + H. This ratio is needed to derive the [D]/[H] ratio from the OH and OD band intensities in comets. From the data of *Engel and Schinke* [1988] and *Shafer et al.* [1989], the photolysis of HDO in the first absorption band favors the production of OD with respect to OH by a factor of 3-4. Nothing is known for photolysis in the second absorption band, which has a similar rate.

H_2O_2 hydrogen peroxide. From the vacuum-ultraviolet absorption data of *Suto and Lee* [1983] and those of *Molina et al.* [1977] in the near ultraviolet and visible, we derive $\beta = 1.34 \times 10^{-4}$ s^{-1}; the contribution of Lyman α is 2.9×10^{-6} s^{-1}. This is in agreement with the determination of *Huebner* [1985] and *Huebner et al.* [1992] based upon older data. Hydrogen peroxide presumably dissociates into $H_2O_2 + h\nu \rightarrow 2$ OH [*Okabe,* 1978].

Hydrocarbons

CH_3CCH propyne (methylacetylene). From the UV absorption spectrum of *Nakayama and Watanabe* [1964] and that recently presented by *Wu et al.* [1992], we evaluate $\beta = 4.2 \times 10^{-5}$ s^{-1} (1.5×10^{-5} s^{-1} from Lyman α).

H_2CCH_2 allene. From UV absorption spectra of *Rabalais et al.* [1971] and *Wu et al.* [1992], we evaluate $\beta = 1.8 \times 10^{-4}$ s^{-1} (1.7×10^{-5} s^{-1} from Lyman α). Propyne and allene were considered by *Krasnopolsky* [1991] among possible parents for the C_3 cometary radical. Very little is known about their photodissociation channels.

C_6H_6 benzene. Benzene is an interesting potential parent molecule because it is the simplest and most volatile PAH. The presence of PAH in comets is strongly suggested by the existence of an emission band at 3.28 mm. We evaluate ultraviolet absorption of C_6H_6 from the oscillator strengths listed by *Leach* [1989], the relative cross sections measured by *Koch and Otto* [1972] and the absolute cross sections of *Person* [1965]. The resulting β is about 1.1×10^{-3} s^{-1} (with a small contribution by Lyman α of only 10^{-5} s^{-1}). It should be noted that the first electronic band X^1A_{1g}-A^1B_{2u} at 263 nm, which has an excitation rate of 1.6×10^{-4} s^{-1}, does not lead to dissociation. The photodissociation of benzene mainly leads to the phenyl radical C_6H_5 and the benzyne radical C_6H_4 [*Yokoyama et al.* 1990]. Further photodissociation of these radicals breaks the aromatic cycle and leads to C_3-bearing radicals.

CO Group: C_3O_2 carbon suboxide

Huntress et al. [1991] give a rate which corresponds to 4.3×10^{-4} s^{-1} at 1 AU, for the solar activity corresponding to the Giotto flyby of P/Halley. From the absorption data of *Roebber et al.* [1967] we roughly estimate $\beta = 1.1 \times 10^{-4}$ s^{-1}, with a contribution of 3.6×10^{-6} s^{-1} for Lyman α. The discrepancy cannot be explained by solar activity alone. Carbon suboxide photodissociation in the near UV gives $CO + C_2O$, C_2O giving in turn C + CO. C_3O_2 was proposed as a source for cometary CO by *Huntress et al.* [1991].

CHO Species

H_2CO formaldehyde. See *Bockelée-Morvan and Crovisier* [1992]. The absorption in the A^1A_2-X^1A_1 band at 260-360 nm is taken from *Rogers* [1990]. Vacuum-ultraviolet absorption is taken from *Mentall et al.* [1971]. The total rate is 1.99×10^{-4} s^{-1}, mostly due to the A^1A_2-X^1A_1 band (the contribution of Lyman α is only 2×10^{-6} s^{-1}). This rate is in good agreement with that of *Huebner and Carpenter* [1979], *Huebner* [1985], and *Huebner et al.* [1992]. A small fraction of the excitation of the A^1A_2-X^1A_1 band yields fluorescence of H_2CO: about 0.3% from the data of *Miller and Lee* [1975]. The main channels of photodissociation are $CO + H_2$, HCO + H and CO + 2 H.

CH_3OH methanol. Bockelée-Morvan et al. [1991 and The rotational lines of methanol in comets, submitted to *Astronomy and Astrophysics*, 1993] derive a rate $\beta = 1.30 \times 10^{-5}$ s^{-1} (including 3.9×10^{-6} s^{-1} from Lyman α) from the absorption data of *Nee et al.* [1985]. This is much smaller than the preceding values, but is in agreement with a recent reevaluation of *Huebner et al.* [1992]. Formaldehyde is assumed to be the dominant photodissociation product of methanol by *Huebner et al.* [1992]. However, *Satyapal et al.* [1989] observed a preferential production of the methoxy radical (CH_3O) from photolysis of CD_3OH at 193 nm. Little is known for photodissociation at shorter wavelengths.

HCOOH formic acid. Laboratory absorption data were published recently by *Suto et al.* [1988*a*]. The corresponding rate is $\beta = 3.25 \times 10^{-5}$ s^{-1} (3.9×10^{-6} s^{-1} due to Lyman α). This is an order of magnitude smaller than previous estimates. Possible dissociation products are HCO + OH and HCOO + H according to *Suto et al.* [1988*a*].

CH_2CO ketene. Laboratory absorption data were published by *Price et al.* [1951] and *Rabalais et al.* [1971]. The corresponding photodestruction rate is $\beta = 4.41 \times 10^{-4}$ s^{-1} (1.2×10^{-5} s^{-1} due to Lyman α). This strong rate is due to the existence of an absorption band in the 350-280 nm region.

CH_3CHO acetaldehyde. Laboratory absorption data in the visible and near UV were recently published by *Martinez et al.* [1992]. Incomplete vacuum-UV data can be found are from *Barnes and Simpson* [1963]. Using the branching ratios to photodissociation compiled by *Atkinson et al.* [1992], the corresponding photodestruction rate is $\beta = 7.5 \times 10^{-5}$ s^{-1} (with a possible contribution of 6×10^{-6} s^{-1} due to Lyman α).

CH_3CH_2OH ethanol. The only laboratory data that could be found are from *Salahub and Sandorfy* [1971]. The corresponding photodestruction rate is $\beta = 1.79 \times 10^{-5}$ s^{-1} (6.9×10^{-6} s^{-1} due to Lyman α). This is not much larger than the rate for methanol.

$(CH_3)_2O$ dimethyl ether. Laboratory absorption data were published recently by *Suto et al.* [1988*b*]. The corresponding photodestruction rate is $\beta = 3.11 \times 10^{-5}$ s^{-1} (6.6×10^{-6} s^{-1} due to Lyman α). (The weak *A-X* electronic band around 235 nm has not been taken into account.)

CH_3COOH acetic acid. Laboratory absorption data were published recently by *Suto et al.* [1988*a*]. The corresponding photodestruction rate is $\beta = 5.07 \times 10^{-5}$ s^{-1} (1.12×10^{-5} s^{-1} due to Lyman α).

CH_3OHCO methyl formate. Laboratory absorption data were published recently by *Suto et al.* [1988*a*]. The corresponding photodestruction rate is $\beta = 4.73 \times 10^{-5}$ s^{-1} (6.9×10^{-6} s^{-1} due to Lyman α).

Nitrogen Compounds

HC_3N cyanoacetylene (propynenitrile). The determinations by *Huebner and Carpenter* [1979] and *Huebner et al.* [1992] were based upon absorption shortwards of 160 nm. *Halpern et al.* [1988] determined that the main photodestructive channel is through $HC_3N + h\nu \rightarrow H + C_3N$, with a threshold at 244 nm. Thus, HC_3N cannot be directly a parent for the cometary CN radical. From the vacuum-UV absorption spectrum of *Connors et al.* [1974] and the near-UV spectrum of *Bruston et al.* [1989], we derive $\beta = 6.6 \times 10^{-5}$ s^{-1} (6×10^{-6} s^{-1} due to Lyman α). This is larger than the previous determinations of Huebner et al., which did not include dissociation through the strong electronic band of HC_3N around 190-230 nm.

Sulphur compounds

H_2S hydrogen sulphide. Crovisier et al. [1991] estimated $\beta = 2.5 \times 10^{-4}$ s^{-1} from the data of *Lee et al.* [1987], with a contribution of 1.5×10^{-5} s^{-1} from Lyman α. This is an order of magnitude less than the previous estimate of *Huebner* [1985] and in near agreement with that of *Huebner et al.* [1992].

OCS carbonyl sulphide. The rates given by *Huebner* [1985] and *Azoulay and Festou* [1986] differed by an order of magnitude. Laboratory absorption data were published by *Rabalais et al.* [1971] from which we derive $\beta = 9.36 \times 10^{-5}$ s^{-1} (4.5×10^{-6} s^{-1} due to Lyman α). This is in agreement with the rates from Azoulay and Festou and *Huebner et al.* [1992]. OCS dissociates almost exclusively into CO + S [*Atkinson et al.,* 1992].

Conclusion

An inspection of Table 1 shows that in many cases, independent determinations of the photodestruction rates are in reasonable agreement. Future laboratory work could further improve these results. One must have in mind, however, that the applicability of these results to cometary conditions is hampered by several problems which do not have obvious solutions. It is therefore illusory to expect an accuracy better than 10-20%.

Unstable Species

In several cases, we are limited by the lack of laboratory data. See Table 1. Some species are unstable in the laboratory (H_2CS, CH_2NH). This is more generally the case for secondary cometary products such as radicals and molecular ions, which have not been included in the table. Almost no reliable laboratory data exist for theses transient species, and rates estimated from ab initio computations are highly uncertain.

Photodissociation Channels and QuantumYields

We wish to know the relative yields for the various possible photodissociation channels. For instance, for the photodissociation of methanol, which is an identified cometary constituent [*Bockelée-Morvan et al.,* 1991], it would be interesting to know the yields for CO and H_2CO, which are two other important cometary molecules. On the other hand, UV absorption does not always yield 100% dissociation and fluorescence may be possible. In our evaluations, as in some of those of *Huebner et al.* [1992], it is assumed that absorption shortwards of the dissociation wavelength threshold was fully dissociative. This may lead to an overestimation of the photodestruction rate. For instance, *Yung et al.* [1984], when modeling the photochemistry of Titan, included a quatum yield in their photodissociation reactions and generally obtained rates smaller than those given in *Huebner et al.* [1992] or in the present work. Precise measurements of the quantum yield for fluorescence and photodissociation are desirable.

Influence of Temperature

Most of the available UV spectra were measured at room temperature. Several efforts were made, however, to obtain spectra at lower temperatures. For instance, *Wu et al.* [1992] measured systematic cross section variations between 300 and 150 K for several hydrocarbons. Such investigations are almost impossible for the less volatile species. Furthermore, cometary molecules are at low rotational temperatures (<100 K) due to adiabatic expansion of the atmosphere and/or spontaneous decay, and far from the conditions where the laboratory spectra were taken. The exact influence of this temperature effect on the evaluations of cometary photorates is still unknown.

Kinematics of the Fragments

We wish to know the distribution of excess energy within electronic, vibrational, rotational and translational energy of the photodissociation fragments. Electronic emission gives prompt emission. Translational energy gives the ejection velocity of the secondary products, and affects their kinematics. Besides the case for water and a few other species, very little is known on this topic.

Anisotropic Ejection of the Fragments

The unidirectional nature of the solar radiation introduces a source of anisotropy. Both theoretical works and laboratory experiments show that in this case, photofragments may not be ejected isotropically in the rest frame of the dissociated molecule [*Okabe,* 1978]. Up to now, this aspect of photodissociation has not been considered in cometary models; it could affect the distribution of secondary products. Laboratory experiments have measured this effect for several molecules under specific conditions, but they do not yet allow us to make quantitative evaluations for comets.

Solar Radiation Field and its Variations

The far- and extreme-UV radiation field of the Sun is varying in the short and long term. Several attempts have been made to relate these variations to activity indicators. The Sun UV spectrum is occasionally monitored from Earth-orbit observatories, but this does not give the ultimate solution, since the comet or the planet under consideration generally does not face the same side of the Sun as the Earth. Most of the photorates variations are due to absorption of the solar Lyman α line. Since, for the molecules discussed above, the photodissociation rates due to Lyman α absorption are given separately, the reader can have an idea of the rates for active Sun by scaling these rates by the relevant Lyman α flux.

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TABLE 1. Photodestruction Rates From Quiet Sun at 1 AU, in s^{-1}

Molecule	J76	HC79	H85	H92	Others		Meudon		Comments
Water group									
H_2O water	5.0×10^{-5}	1.2×10^{-5}	1.2×10^{-5}	1.2×10^{-5}			1.3×10^{-5}	C89	
HDO deuterated water							1.3×10^{-5}	C92	
H_2O_2 hydrogen peroxide			1.4×10^{-4}	1.4×10^{-4}			1.3×10^{-4}	C92	
Hydrocarbons									
CH_4 methane	8.3×10^{-6}	7.7×10^{-6}	7.8×10^{-6}	7.6×10^{-6}	8.0×10^{-6}	A87			
C_2H_2 acetylene	1.7×10^{-4}	3.1×10^{-5}	1.4×10^{-5}	1.4×10^{-5}					
C_2H_4 ethylene		4.8×10^{-5}	4.8×10^{-5}	4.8×10^{-5}					
C_2H_6 ethane			1.1×10^{-5}	1.1×10^{-5}					
CH_3CCH propyne	2.0×10^{-4}				3.5×10^{-5}	K91	4.2×10^{-5}	C92	
CH_2CCH_2 allene					1.4×10^{-4}	K91	1.8×10^{-4}	C92	
C_6H_6 benzene							1.1×10^{-3}	C92	
CO group									
CO carbon monoxide		6.5×10^{-7}	6.5×10^{-7}	7.5×10^{-7}					
CO_2 carbon dioxide		2.0×10^{-6}	2.0×10^{-6}	2.0×10^{-6}					
C_3O_2 carbon suboxide					4.3×10^{-4}	HA91	1.1×10^{-4}	C92	
CHO species									
H_2CO formaldehyde	5.3×10^{-4}	2.8×10^{-4}	2.8×10^{-4}	2.8×10^{-4}	2.0×10^{-4}	M93	2.0×10^{-4}	BC92	
CH_3OH methanol	4.8×10^{-5}	2.7×10^{-4}	2.7×10^{-4}	1.2×10^{-5}			1.3×10^{-5}	BC91	
HCOOH formic acid	1.4×10^{-4}	7.6×10^{-4}	8.8×10^{-4}	8.8×10^{-4}			3.2×10^{-5}	C92	
CH_2CO ketene							4.4×10^{-4}	C92	
CH_3CHO acetaldehyde	6.2×10^{-4}		5.5×10^{-5}	5.7×10^{-5}			7.5×10^{-5}	C92	
C_2H_5OH ethanol							1.8×10^{-5}	C92	
$(CH_3)_2O$ dimethyl ether	1.2×10^{-4}						3.1×10^{-5}	C92	
CH_3COOH acetic acid							5.1×10^{-5}	C92	
CH_3OHCO methyl formate							4.7×10^{-5}	C92	
HC_3HO propynal					1.5×10^{-3}	K91			
Nitrogen compounds									
NH_3 ammonia	4.8×10^{-4}	1.8×10^{-4}	1.8×10^{-4}	1.8×10^{-4}	1.5×10^{-4}	A87			
HCN hydrogen cyanide	1.1×10^{-5}	1.3×10^{-5}	1.3×10^{-5}	1.3×10^{-5}			1.5×10^{-5}	BC85	
HC_3N cyanoacetylene	7.7×10^{-5}	2.8×10^{-5}	2.8×10^{-5}	3.9×10^{-5}	3.4×10^{-5}	K91	6.6×10^{-5}	C92	
HC_5N cyanodiacetylene									no lab. data
CH_3CN methyl cyanide							6.7×10^{-6}	BC85	
CH_2NH methanimine									no lab. data
CH_3NH_2 methylamine	6.7×10^{-4}								
HNCO isocyanic acid	7.7×10^{-5}		2.9×10^{-5}	2.9×10^{-5}					
NH_2CHO formamide	6.7×10^{-4}								
C_2N_2 dicyanogen	9.1×10^{-5}						3.1×10^{-5}	BC85	
Sulphur compounds									
H_2S hydrogen sulphide			3.3×10^{-3}	3.2×10^{-4}			2.5×10^{-4}	CD91	
CS_2 carbon disulphide			3.5×10^{-3}	2.9×10^{-3}	1.7×10^{-3}	JB86			
SO sulphur monoxide				6.2×10^{-4}	1.5×10^{-4}	KA91			
SO_2 sulphur dioxide			2.5×10^{-4}	2.1×10^{-4}	2.1×10^{-4}	KA91			
OCS carbonyl sulphide			6.5×10^{-4}	1.0×10^{-4}	8.0×10^{-5}	AF86	9.4×10^{-5}	C92	
H_2CS thioformaldehyde									no lab. data

A87 [*Allen et al.,* 1987], AF86 [*Azoulay and Festou,* 1986], BC85 [*Bockelée-Morvan and Crovisier,* 1985], BC91 [*Bockelée-Morvan et al.,* 1991], BC92 [*Bockelée-Morvan and Crovisier,* 1992], C89 [*Crovisier,* 1989], C92 (this work), CD91 [*Crovisier et al.,* 1991], H85 [*Huebner,* 1985], H92 [*Huebner et al.,* 1992], HA91 [*Huntress et al.,* 1991], HC79 [*Huebner and Carpenter,* 1979], J76 [*Jackson,* 1976], JB86 [*Jackson et al.,* 1986], K91 [*Krasnopolsky,* 1991], KA91 [*Kim and A'Hearn,* 1991], M93 [*Meier et al.,* 1993].