

Statistical universal branching ratios for cosmic ray dissociation, photodissociation, and dissociative recombination of the $C_{n=2-10}$, $C_{n=2-4}H$ and C_3H_2 neutral and cationic species

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ABSTRACT

Context. Fragmentation-branching ratios of electronically excited molecular species are of first importance for the modeling of gas phase interstellar chemistry. Despite experimental and theoretical efforts that have been done during the last two decades there is still a strong lack of detailed information on those quantities for many molecules such as C_n , C_nH or C_3H_2 .

Aims. Our aim is to provide astrochemical databases with more realistic branching ratios for C_n ($n = 2$ to 10), C_nH ($n = 2$ to 4), and C_3H_2 molecules that are electronically excited either by dissociative recombination, photodissociation, or cosmic ray processes, when no detailed calculations or measurements exist in literature.

Methods. High velocity collision in an inverse kinematics scheme was used to measure the complete fragmentation pattern of electronically excited C_n ($n = 2$ to 10), C_nH ($n = 2$ to 4), and C_3H_2 molecules. Branching ratios of dissociation were deduced from those experiments. The full set of branching ratios was used as a new input in chemical models and branching ratio modification effects observed in astrochemical networks that describe the dense cold Taurus Molecular Cloud-1 and the photon dominated Horse Head region.

Results. The comparison between the branching ratios obtained in this work and other types of experiments showed a good agreement. It was interpreted as the signature of a statistical behavior of the fragmentation. The branching ratios we obtained lead to an increase of the C_3 production together with a larger dispersion of the daughter fragments. The introduction of these new values in the photon dominated region model of the Horse Head nebula increases the abundance of C_3 and C_3H , but reduces the abundances of the larger C_n and hydrocarbons at a visual extinction A_V smaller than 4.

Conclusions. We recommend astrochemists to use these new branching ratios. The data published here have been added to the online database KIDA (Kinetic Database for Astrochemistry, <http://kida.obs.u-bordeaux1.fr>).

Key words. astrochemistry – molecular data – ISM: clouds – photon-dominated region (PDR)

1. Introduction

Carbon clusters and highly unsaturated hydrocarbons are among the molecules most often observed in the Inter Stellar Medium (ISM) (<http://astrochemistry.net>). They are present in diffuse clouds (Hobbs & Campbell 1982; Maier et al. 2001; Galazutdinov et al. 2002; Roueff et al. 2002), in dense clouds (Bell et al. 1999; Fossé et al. 2001; Dickens et al. 2000), in circumstellar envelopes (Cernicharo et al. 2002), protostar envelopes (Sakai et al. 2008), planetary nebula (Cernicharo et al. 2001) and in photon dominated regions (PDR) (Teyssier et al. 2004). They are also present around evolved (carbon) stars (Hinkle et al. 1988), in the Titan ionosphere (Lavvas et al. 2008) or in comet comae (Helbert et al. 2005). Since their discovery in the early 70s, many studies have been devoted to those species (Van Orden & Saykally 1998), mostly on their structural and

spectroscopic properties. The spectroscopic studies were pushed mainly by the dramatically increasing performances of the observation capabilities, while for the structural studies it was also driven by the increasing use of carbon-based materials.

The origin of carbon clusters and highly unsaturated hydrocarbons and their abundances in the ISM objects is still a puzzling question. It is thought that they may come from gas-phase synthesis as well as be released into the gas-phase of carbonaceous solid materials that are present in the ISM. Therefore the detailed investigation of their origin and abundances requires homogeneous and heterogeneous chemical reactions to be explored. Whatever the carbon reservoir, those molecules are processed in the gas phase by neutral-neutral and ion-neutral collisions as well as cosmic rays (CR), ultra violet (UV) photons and dissociative recombination (DR) from collisions with the thermal electrons. These last three mechanisms lead to highly

electronically excited species that may undergo fragmentation. The detailed dynamics and the fragmentation channels have thus to be investigated to enable their incorporation into various astrochemical models.

In gas phase chemistry, chemical networks for carbon clusters and highly unsaturated hydrocarbons synthesis have been proposed for a large variety of astrophysical conditions (Greenberg 1976; Black & Dalgarno 1977; Herbst & Leung 1989; Jura & Kroto 1990; Millar et al. 2000; Turner et al. 2000; Röllig et al. 2007). Nevertheless, despite the strong theoretical and experimental efforts in structure and spectroscopy, many of the reaction rates still remain uncertain and may consequently limit the confidence in chemical models.

It is convenient to write the reaction rate coefficient for a reaction $A+B \rightarrow C+D$ as the product of a total reaction rate coefficient k_{tot} and a branching ratio factor (BR)

$$k[A+B \rightarrow C+D] = k_{\text{tot}}[A+B \rightarrow (AB)^*] \times \text{BR}[(AB)^* \rightarrow C+D]. \quad (1)$$

The reliability of the reaction rate may be improved when the total rate and BR are independently predicted. Most of the reaction rates are sensitive to the surrounding medium (temperature and photon spectrum for instance). In Eq. (1) this dependency is implicitly included in the total rate, whereas BRs are assumed to be constant. This assumption of constant BRs may however be inappropriate for some reactions as for example, neutral-neutral or ion-neutral reactive collisions where energy barriers can be present. For electronic excitation mechanisms followed by dissociation that occur in the ISM, the intermediate complex AB is appropriately described by an ensemble of molecules prepared in various highly excited electronic states. It has been pointed out several times that this physical situation fits the requirements of statistical theory concepts very well (Herbst 1978; Leger et al. 1989; Bettens & Herbst 1995). In the particular case of small species, non-statistical mechanisms need also to be considered because the density of states is generally not high enough, even if high-energy excitations are involved. Non-statistical behavior may also arise in rare photodissociation situations, where a dissociative state is resonant with a discrete line from a local photon source (van Dishoeck 1988).

In the first part of this paper we will report on measured BRs after electronic excitations that take place in a high-velocity collision experiment. In the second part we will introduce through the comparison between the different processes that are responsible for the electronic excitations in the ISM, the idea of statistical universal BR for cosmic-ray-induced dissociation, photodissociation, and dissociative recombination processes. We will then propose to correct BRs of current online databases (OSU¹, UMIST²) when they are not resulting from measurements or detailed calculations. In a last part we will observe the influence of the new branching ratios on two typical astrochemical models that are aimed to simulate the dense clouds and the Horse Head PDR.

2. Fragmentation of $C_{n=2-10}$, $C_{n=2-4}H$ and C_3H_2 molecules that are electronically excited by high-velocity collision

2.1. Experimental set-up

The experimental set-up has already been described in detail in previous papers (Chabot et al. 2002; Tuna et al. 2008) and only

¹ <http://www.physics.ohio-state.edu/~eric/research.html>

² <http://www.udfa.net/>

a brief description is given here. Tandem MP (15 MVolts) accelerator at the Institut de Physique Nucléaire d'Orsay produced molecular cationic beams of several MeV of energy (Waast et al. 1996). After magnetic analysis and collimation, singly charged molecules collided at high velocity (few a.u.) with a single helium target atom. Half a meter downstream, a parallel-plates electrostatic deflector analyzed parents and fragments with respect to their q/m ratio. A set of silicon detectors intercepted all the trajectories in a dedicated chamber. It is because of their high velocity that parent and fragments may be detected with silicon detectors. This type of detector enables the measurement of the kinetic energy of the particle, i.e., with high-energy molecular beams of constant velocity, the mass of the detected molecule. Moreover, silicon detectors are 100% efficient and, owing to the kinematics, small detector sizes can cover 100% of the solid angle for fragments emission. All the detectors are operating in coincidence, event by event. To get branching ratios on neutral species, the grid technique (Morgan et al. 1971; Larsson 1995) may be used when the number of channels is small. In this method, a grid of known transmission is placed in front of the detector. Branching ratios of dissociation are linked to the recorded mass spectra through a set of linear equations. In the present work, because high-energy molecular beams were used, the shape of the current signal from silicon detectors could be analyzed to resolve a pile-up of several neutral fragments (Chabot et al. 2002). However, this technique was insufficient to get the whole information for the hydrogenated species. The grid technique was then mixed with a signal shape analysis to finally fully resolve the fragmentation of neutral and cationic C_n , C_nH , and C_3H_2 species (Tuna et al. 2008).

2.2. High-velocity collision (HVC) processes

2.2.1. The excitation processes

During the fast ($\sim 10^{-16}$ s) collision between a molecule X and an atom, charge transfer (HVC - CT) may occur:



Owing to the high initial velocity of the transferred electron, electronically excited states as well as the ground state are populated (Chabot et al. 2006). Until now no calculations have been performed to predict the internal energy distribution associated with this process. Nevertheless, internal energy distributions may be deduced for the present species from experimental multiplicity distribution, i.e. probabilities associated with a given number of emitted fragments (Tuna et al. 2008). Some internal energy distributions are reported in Fig. 1.

In addition excitation (HVC - E) may also occur during the collision.



Let us note in this velocity regime that only electronic excitations take place (Wohrer et al. 1998). Calculations of internal energy following HVC-E have been performed through independent atom and electron model (Wohrer & Watson 1993) using doubly differential probabilities with the energy and impact parameter calculated within the classical trajectory monte carlo (CTMC) theory (Maynard 1999, priv.comm.) These calculated distributions agree very well with the energy distributions shown in Fig. 2, which are deduced from experimental multiplicity distributions. Ionization is also occurring in HVC. In the present experiments with cationic beams it leads to multiply charged species which are generally also electronically excited

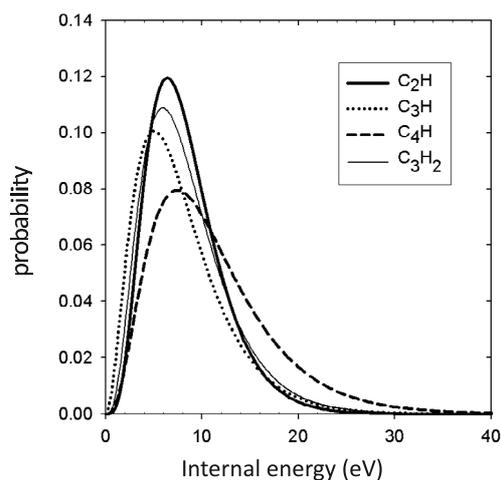


Fig. 1. Internal energy distributions following HVC-charge transfer (see text). Thick solid line: C_2H , dotted line: C_3H , broken line: C_4H , thin solid line: C_3H_2 . Peak energies and widths of distributions may be regarded with errors on the order of a few eV. Details on the method of extraction are given in Tuna et al. (2008).

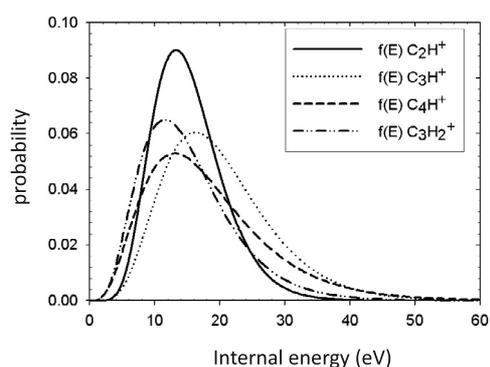


Fig. 2. Internal energy distributions following HVC-excitation (see text). Solid line: C_2H^+ , dotted line: C_3H^+ , broken line: C_4H^+ , dot-dashed line: $C_3H_2^+$. Peak energies and widths of distributions may be regarded with errors on the order of a few eV. Details on the method of extraction are given in Tuna et al. (2008).

(Chabot et al. 2010). We will not report on the fragmentation of these multiply charged species, because this is out of the scope of this paper.

2.2.2. Fragmentation of electronically excited species by high-velocity collision

Statistical hypothesis has been proposed and often applied to calculate the fragmentation of finite size systems. It stipulates that all accessible micro-states are equiprobably populated, and conservations of energy and momentum define which these micro-states are. Their ensemble is forming a so-called phase space. Theoretical predictions have to rely on numeration of individual states or integration in the phase-space. Two physical situations are presumably close to the statistical behavior. In the first one, the system gets enough time to explore all the phase space whatever the bottlenecks in this random exploration. In the second physical situation, the system has no time to explore the whole phase space, but is prepared in such a large set of initial states that sampling of the phase space is presumably achieved by the

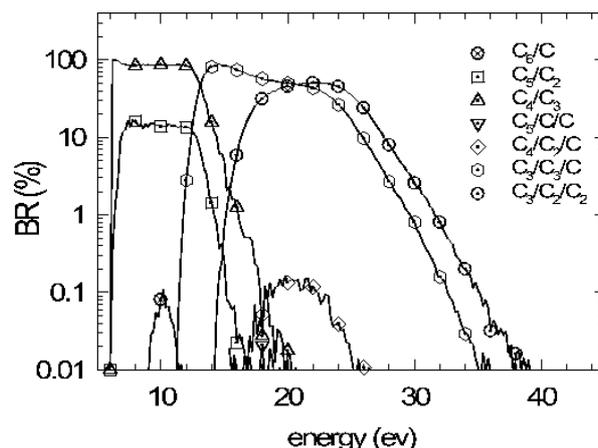


Fig. 3. Theoretical fragmentation BRs as a function of internal excitation energy for a C_7 carbon cluster. Calculations were performed with the MMMC statistical theory (Diaz-Tendero et al. 2006).

Table 1. Probability of dissociation of C_3H_2 produced in HVC-CT reaction as a function of number of the emitted fragments (N_f).

N_f	Proba (\pm err)
1	0.16 (0.05)
2	0.43 (0.07)
3	0.28 (0.08)
4	0.08 (0.03)
5	0.05 (0.01)

Notes. $N_f = 1$ corresponds to non-dissociative HVC-CT.

entrance channels. Fragmentation of molecules that are electronically excited by HVC processes are clearly related to the second scenario and therefore can generally be interpreted in the frame of a statistical theory. For example, a statistical theory of fragmentation, the microcanonical metropolis monte carlo (MMMC) (Gross & Hervieux 1995), has been applied to the carbon clusters C_n and found to agree very well with the experimental measurements (Martinet et al. 2004).

For C_n , C_nH , and C_3H_2 the creation of each fragment costs roughly the same amount of energy (4–7 eV³). Therefore the fragmentation BRs exhibit specific energy domains associated to a given multiplicity (see Fig. 3). Normalizing the branching ratios to multiplicity probabilities somehow vanishes the role of the input energy on BRs. To illustrate this point, we present in Fig. 4 the calculated BRs for C_7 . They were obtained by convolution of the theoretical curves of the Fig. 3 with Gaussian shapes centered at energies from 0 to 15 eV. The variations are found to be small, i.e. within $\pm 10\%$. It shows that as long as phase space scrambling is achieved, normalized BRs are mostly insensitive to the internal energy distribution, unlike the multiplicity distributions.

2.3. Experimental results and analysis

Results for C_3H_2 fragmentation (multiplicity distribution: Table 1, BR: Fig. 5) and $C_3H_2^+$ (multiplicity distribution: Table 2, BR: Fig. 6) have been recently obtained. Detailed physical discussion on this species will be done elsewhere. Note that the detachment of one or two hydrogens is the most favorable channel. Fragmentation results for C_n and C_nH species can be found

³ C_3H_2 formation energy is only 3 eV, but there is a barrier close to 6 eV (Leonori et al. 2008).

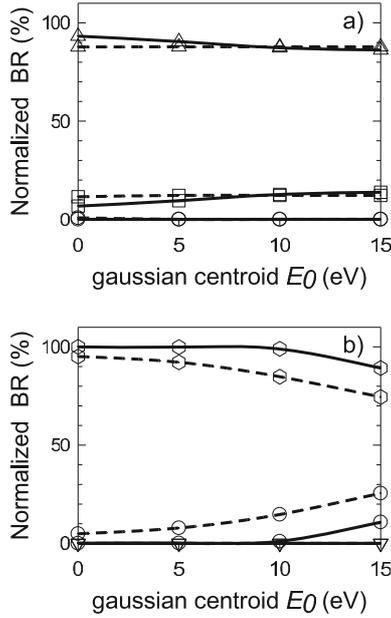


Fig. 4. C_7 calculated BRs with various internal energy distribution. The BRs were obtained by convolution of theoretical curves of Fig. 3 with the gaussian energy distribution: $p(E) = \frac{1}{\sigma\sqrt{2\pi}} \exp(-\frac{1}{2}(\frac{E-E_0}{\sigma})^2)$. The BRs are normalized to the multiplicity probability. Solid lines: $\sigma = 2$ eV dotted lines: $\sigma = 5$ eV. **a)** BR for two fragments breakup: triangles up: C_4/C_3 , squares: C_5/C_2 , circle: C_6/C . **b)** BR for three fragments breakup: hexagons: $C_3/C_3/C$, circle: $C_3/C_2/C_2$, triangle down: $C_4/C_2/C$ and $C_5/C/C$.

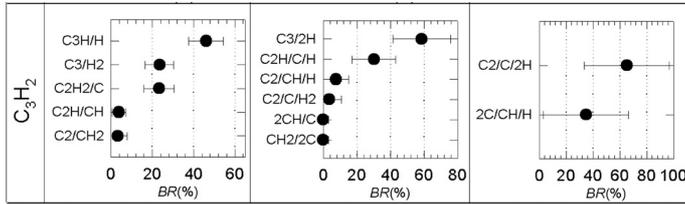


Fig. 5. Experimental branching ratios of neutral C_3H_2 produced in HVC-CT reaction at a fixed number of emitted fragments (N_f) from left to right ($N_f = 2, 3, 4$).

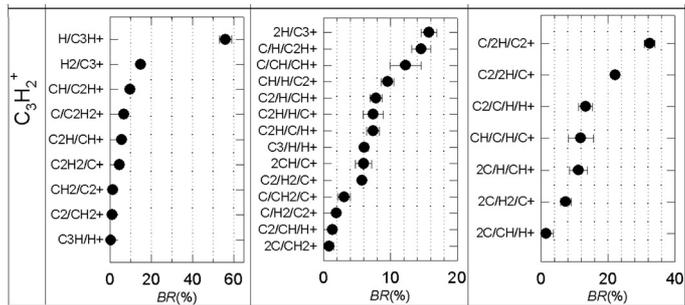


Fig. 6. Experimental branching ratios of $C_3H_2^+$ produced in HVC-E reaction at a fixed number of emitted fragments (N_f) from left to right ($N_f = 2, 3, 4$).

elsewhere (Martinet et al. 2004; Díaz-Tendero et al. 2006; Tuna et al. 2008). A subset of these results are shown in Figs. 7 to 9. Briefly, the most favorable channels were found to be H production for C_nH and C_3 production for C_n . These channels are indeed the most exothermic ones. For C_n , C_3 is a magic number because of the shell closure (Van Orden & Saykally 1998).

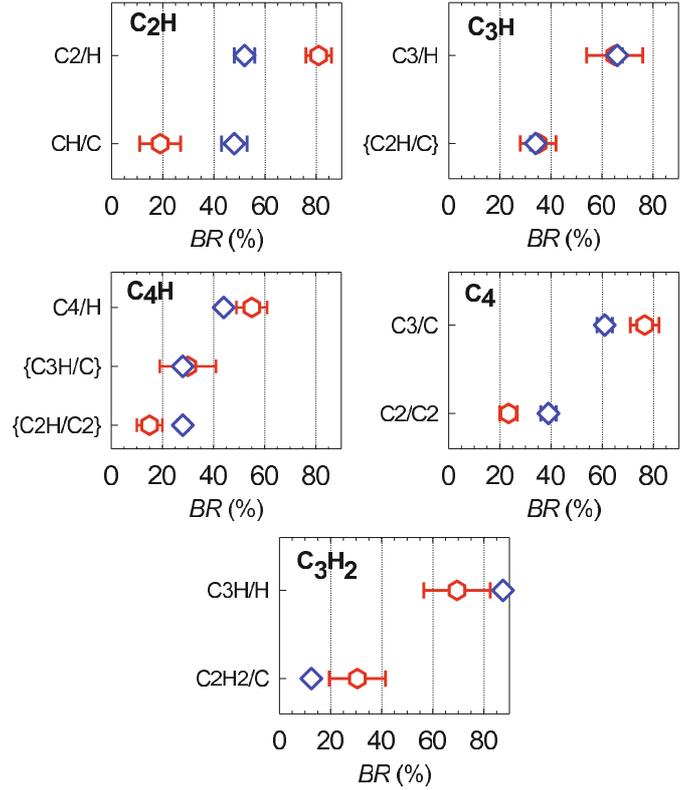


Fig. 7. Comparison between a two fragments breakup after dissociative recombination (DR) (blue diamonds) and HVC-CT (red hexagons – this work). DR-BR are adapted from Ehlerding et al. (2004) for C_2H ; Angelova et al. (2004b) for C_3H , C_3H_2 ; Angelova et al. (2004a) for C_4H and Heber et al. (2006) for C_4 . The parenthesis on Y-axis labels mean that hydrogen may be localized on either of the two fragments in DR experiments.

Table 2. Probability of dissociation of $(C_3H_2^+)^*$ produced in HVC-E reaction as a function of number of the emitted fragments (N_f).

N_f	Proba (\pm err)
2	0.34 (0.02)
3	0.43 (0.03)
4	0.19 (0.08)
5	0.05 (0.01)

Notes. Non-dissociative excited species are not detected in the experiment.

3. Application of HVC fragmentation branching ratios in astrochemical reaction networks

3.1. Statistical fragmentation relevance in the context of ISM chemistry

Three physical processes are involved in the creation of transient molecular electronically excited species in ISM (Duley & Williams 1984). They are HVC with cosmic rays and secondary electrons (Spitzer & Tomasko 1968), photoabsorption in the various local radiation fields (Duley & Williams 1984), and recombination between molecular ions and thermal electrons in the local plasma (Herbst 2003). The two latter processes are generally dominating, except perhaps in dark clouds where CR-induced secondary electrons may have a significant contribution (Prasad & Tarafdar 1983; Gredel et al. 1987).

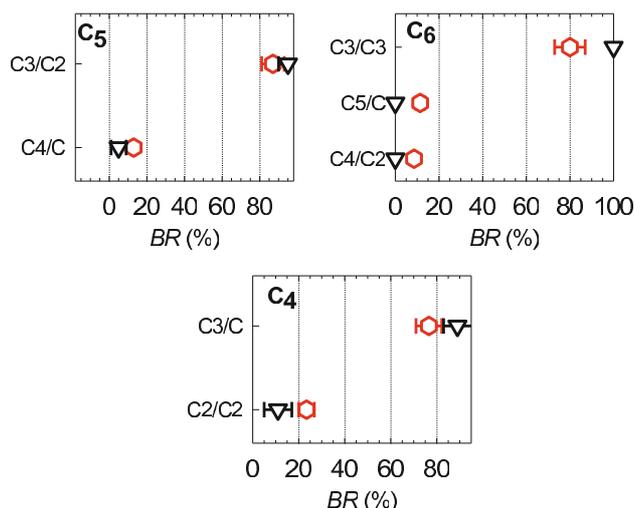


Fig. 8. Comparison between a two fragments-breakup after photodissociation (black triangles) and HVC-CT (red hexagons – this work). The BRs for photodissociation are adapted from Choi et al. (2000) measurements.

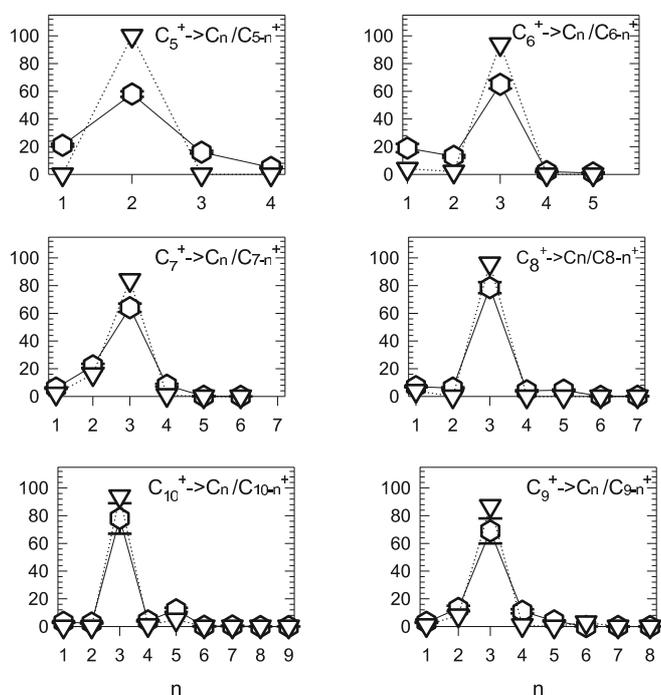


Fig. 9. Comparison between a two fragments-breakup after photodissociation (triangles) and HVC-E (hexagons – this work). The BRs for photodissociation are adapted from Geusic et al. (1986).

Because of the energy and momentum conservation laws the internal energy, in the electronic recombination process, should be close to the neutral species ionization potential (IP). In hydrocarbon molecules, the IP is always higher than the dissociation energy (DE). Then, fragmentation will occur rapidly for small size molecules, as compared to radiative deexcitation. According to the IP values (8 to 12 eV; Van Orden & Saykally 1998) and DE (4 to 7 eV; Díaz-Tendero et al. 2006; Tuna et al. 2008), the channels that lead to two fragments are expected to be the most populated.

Statistical fragmentation may be invoked in the dissociative recombination process because there are always many

electronically excited states close to the IP in carbon and hydrocarbon molecules (van Hemert & van Dishoeck 2008). As a result a rather efficient sampling of the phase space should be performed in the entrance channels, and dissociative recombination BRs should be governed by a statistical fragmentation as in HVC. Figure 7 shows a comparison between HVC-CT and DR results for C₄, C₂H, C₃H, C₄H, and C₃H₂. Some of those results (C₃H and C₄) are not BR but a summation of BRs. Indeed, a resolution of C-H bond breaking is not always achieved in DR experiments nearby storage ring facilities due to the limitation of the grid method (see experimental section). The DR data agree with HVC-CT data within ±10% on average. This small discrepancy could arise because the internal energy distributions are quite different between HVC-CT (large distributions) and DR (narrow distribution around IP). An exception is seen in C₂H, where the agreement is poor. It may come from a non-statistical behavior of the fragmentation for a triatomic molecule this small.

In the ISM, molecules are photo-dissociated and photo-ionized because of the absorption of UV photons. These UV photons are produced by nearby stars. The interstellar standard radiation field (ISRF) embedding the gas in the Galaxy has been determined by Draine (Draine 1978). In star-forming regions, the UV flux is the sum of the ISRF plus the emission spectra of the nearby stars. UV photons are absorbed in the continuum by dust and via discrete lines of the most important molecules. Photo-absorption rates thus depend on the optical depth. In the interior of dense clouds, UV photons are produced by the excitation in Lyman and Werner electronic states of molecular hydrogen by CR-secondary electrons (Prasad & Tarafdar 1983).

Taking into account the strong density of electronically excited states for the carbon and hydrocarbon molecules in the 6–11 eV energy range (van Hemert & van Dishoeck 2008), many electronic transitions are expected to occur, resulting in a wide distribution of prepared excited states. Then fragmentation, as for HVC and DR, is expected to be governed by statistical behavior. Figure 8 presents fragmentation BR for C₄, C₅, and C₆ carbon clusters obtained by photodissociation and those from HVC-CT. In the reported photodissociation experimental results (Choi et al. 2000), the incident photon energy has been varied up to 6 eV with some contribution of multi-photon absorption. The horizontal bars on these measurements correspond to a variation of the BRs with the different photon energies. One observes agreement within ±10% between photodissociation and HVC-CT BR. Note that these photodissociation experiments were favorably compared to statistical calculations (Choi et al. 2000). Figure 9 presents the same comparison for cationic C_n⁺ (n = 5 to 10) species. Photodissociation was produced by single and runaway multiphoton absorption (Sowa et al. 1991; Geusic et al. 1986). Therefore, those results have to be considered with caution. Inside the above restrictions, the BRs from photodissociation and HVC-E processes still agree also quite well.

In view of the above comparisons, a universality of statistical BRs may be proposed. In order to take into account the particularity of the different electronic processes, an error bar (1σ) of ±10% seems reasonable to apply.

3.2. New BRs for astrochemistry databases

We propose here, to use the present complete set of BR obtained with HVC as statistical universal BRs (HVC-SUBR) for C_n, C_nH, and C₃H₂ when they are missing or guessed in ISM databases. Table 3 presents CR HVC-SUBR for C_n, C_nH, and C₃H₂ together with OSU-01-2007 CR-BR. We do not report the

Table 3. New HVC-SUBR for CR reactions.

Reactants	Total rate (10^3 s^{-1}) (OSU-01-2007)	Products	HVC-SUBR	OSU-01-2007-BR
$\text{C}_2\text{H} + \text{cr}$	5.0	$\text{C}_2 + \text{H}$	0.81	1.00
		$\text{C} + \text{CH}$	0.19	0.00
$\text{C}_3\text{H} + \text{cr}$	5.0	$\text{C}_3 + \text{H}$	0.65	1.00
		$\text{C}_2\text{H} + \text{C}$	0.33	0.00
		$\text{C}_2 + \text{CH}$	0.02	0.00
$\text{C}_4 + \text{cr}$	1.0	$\text{C}_3 + \text{C}$	0.77	1.00
		$\text{C}_2 + \text{C}_2$	0.23	0.00
$\text{C}_3\text{H}_2 + \text{cr}$	5.0	$\text{C}_3\text{H} + \text{H}$	0.46	1.00
		$\text{C}_3 + \text{H}_2$	0.24	0.00
		$\text{C}_2\text{H}_2 + \text{C}$	0.23	0.00
		$\text{C}_2\text{H} + \text{CH}$	0.04	0.00
		$\text{C}_2 + \text{CH}_2$	0.03	0.00
$\text{C}_4\text{H} + \text{cr}$	5.00	$\text{C}_4 + \text{H}$	0.58	1.00
		$\text{C}_3\text{H} + \text{C}$	0.26	0.00
		$\text{C}_3 + \text{CH}$	0.00	0.00
		$\text{C}_2\text{H} + \text{C}_2$	0.16	0.00
$\text{C}_5 + \text{cr}$	1.00	$\text{C}_4 + \text{C}$	0.13	1.00
		$\text{C}_3 + \text{C}_2$	0.87	0.00
$\text{C}_6 + \text{cr}$	1.00	$\text{C}_5 + \text{C}$	0.09	1.00
		$\text{C}_4 + \text{C}_2$	0.11	0.00
		$\text{C}_3 + \text{C}_3$	0.80	0.00
$\text{C}_7 + \text{cr}$	1.00	$\text{C}_6 + \text{C}$	0.01	1.00
		$\text{C}_5 + \text{C}_2$	0.19	0.00
		$\text{C}_4 + \text{C}_3$	0.80	0.00
$\text{C}_8 + \text{cr}$	1.00	$\text{C}_7 + \text{C}$	0.03	1.00
		$\text{C}_6 + \text{C}_2$	0.01	0.00
		$\text{C}_5 + \text{C}_3$	0.90	0.00
		$\text{C}_4 + \text{C}_4$	0.06	0.00
$\text{C}_9 + \text{cr}$	1.00	$\text{C}_8 + \text{C}$	0.00	1.00
		$\text{C}_7 + \text{C}_2$	0.06	0.00
		$\text{C}_6 + \text{C}_3$	0.66	0.00
		$\text{C}_5 + \text{C}_4$	0.28	0.00

Notes. Total rates and BR of OSU-01-2007 (<http://www.eric/o.html>) are also reported. We do not report on C_{10} because this reaction is not included in OSU-01-2007.

Umist data base values because the BR are almost the same as those of OSU-01-2007. When reactions were not considered in OSU-01-2007, we do however used Umist06 rates. Note that the most recent version of the osu database (USU.2009) has the same BR as OSU-01-2007 for these species. All CR-BR from OSU-01-2007 result from the pioneering estimates (Leung et al. 1984). In most cases, they used zero level statistical behavior to predict BR, i.e. all the dissociation goes only to the most exothermic channel. It is consistent with the HVC-SUBR obtained for the C_nH , but not for the C_n species. Indeed, they assumed C_{n-1}/C and $\text{C}_{n-1}/\text{C}_2$ to be the most exothermic channels, whereas it is always the $\text{C}_{n-3}/\text{C}_3$ channel. In general, HVC-SUBR lead to a much wider dispersion of the fragmentation channels. It is remarkable that C_3 production by CR is strongly enhanced.

Table 4 presents new DR HVC-SUBR for C_n^+ , C_nH^+ , and C_3H_2^+ together with OSU-01-2007 BR and reaction rates. We do not report on C_2H^+ and C_4^+ because OSU-01-2007 used experimental DR-BR (Ehlerding et al. 2004; Heber et al. 2006). For C_3H^+ , in spite of existing partial measurements (Angelova et al. 2004b), the OSU-01-2007 database uses the same probability for the emission of C or H. Although Leung et al. (1984) assumed 100% of H emission, equiprobability was proposed by Millar et al. (1988), based on more detailed calculations. The HVC-SUBR are indeed between these two extreme situations. For

Table 4. New HVC-SUBR for DR reactions.

Reactants	Total rate ($10^{-7} \text{ cm}^3 \text{ s}^{-1}$) (OSU-01-2007)	Products	HVC-SUBR	OSU-01-2007-BR
$\text{C}_3\text{H}^+ + \text{e}^-$	3.0	$\text{C}_3 + \text{H}$	0.65	0.50
		$\text{C}_2\text{H} + \text{C}$	0.33	0.50
		$\text{C}_2 + \text{CH}$	0.02	0.00
$\text{C}_3\text{H}_2^+ + \text{e}^-$	3.6	$\text{C}_3\text{H} + \text{H}$	0.46	0.42
		$\text{C}_3 + \text{H}_2$	0.23	0.42
		$\text{C}_2\text{H}_2 + \text{C}$	0.24	0.08
		$\text{C}_2\text{H} + \text{CH}$	0.04	0.00
		$\text{C}_2 + \text{CH}_2$	0.04	0.08
$\text{C}_4\text{H}^+ + \text{e}^-$	3.00	$\text{C}_4 + \text{H}$	0.58	0.40
		$\text{C}_3\text{H} + \text{C}$	0.26	0.15
		$\text{C}_3 + \text{CH}$	0.00	0.15
		$\text{C}_2\text{H} + \text{C}_2$	0.16	0.30
$\text{C}_5^+ + \text{e}^-$	3.00	$\text{C}_4 + \text{C}$	0.13	0.50
		$\text{C}_3 + \text{C}_2$	0.87	0.50
$\text{C}_6^+ + \text{e}^-$	20.00	$\text{C}_5 + \text{C}$	0.09	0.50
		$\text{C}_4 + \text{C}_2$	0.11	0.50
		$\text{C}_3 + \text{C}_3$	0.80	0.00
$\text{C}_7^+ + \text{e}^-$	23.00	$\text{C}_6 + \text{C}$	0.01	0.43
		$\text{C}_5 + \text{C}_2$	0.19	0.43
		$\text{C}_4 + \text{C}_3$	0.80	0.14
$\text{C}_8^+ + \text{e}^-$	20.00	$\text{C}_7 + \text{C}$	0.03	0.50
		$\text{C}_6 + \text{C}_2$	0.01	0.50
		$\text{C}_5 + \text{C}_3$	0.90	0.00
		$\text{C}_4 + \text{C}_4$	0.06	0.00
$\text{C}_9^+ + \text{e}^-$	20.00	$\text{C}_8 + \text{C}$	0.00	0.50
		$\text{C}_7 + \text{C}_2$	0.06	0.50
		$\text{C}_6 + \text{C}_3$	0.66	0.00
		$\text{C}_4 + \text{C}_4$	0.28	0.00
$\text{C}_{10}^+ + \text{e}^-$	20.00	$\text{C}_9 + \text{C}$	0.01	0.50
		$\text{C}_8 + \text{C}_2$	0.01	0.50
		$\text{C}_7 + \text{C}_3$	0.70	0.00
		$\text{C}_6 + \text{C}_4$	0.03	0.00
		$\text{C}_5 + \text{C}_5$	0.25	0.00

Notes. Total rates and BR of OSU-01-2007 (<http://www.eric/o.html>) are also reported. We do not report about DR on C_2H^+ and C_4^+ because OSU-01-2007 used experimental measurements.

C_4H^+ , OSU-01-2007 uses the experimental DR- BR (Angelova et al. 2004a) for the breaking of the C-C bonds and, without any available information on the ratio between emission of C or CH, OSU-01-2007 took it equal to 1. The HVC-SUBR results show that actually CH emission is very unlikely. For C_3H_2^+ , OSU-01-2007 uses the experimental DR-BR (Angelova et al. 2004b) for the C-C bonds. For the missing experimental information on the proportion between H and H_2 emission when the carbon skeleton stays intact and the proportion between C and CH when it is broken, OSU-01-2007 still assumed equal probability. As for C_3H^+ , HVC-SUBR show that CH emission is unlikely. For the proportion of evaporation of H and H_2 , equal probability is not too far from HVC-SUBR. It is remarkable that the HVC-SUBR give a ratio H_2/H (35%) in qualitative agreement with the recent experiment of neutral-neutral reaction $\text{C} + \text{C}_2\text{H}_2 \rightarrow \text{C}_3\text{H}_2^+$ (Leonori et al. 2008). For the C_n^+ species, as for CR, note that HVC-SUBR predict an enhanced production of C_3 driven by DR together with a more extended dispersion in the carbon cluster-mass daughters compared to OSU-01-2007.

Table 5 presents a new photon HVC-SUBR for C_n , C_nH , and C_3H_2 together with OSU-01-2007 BR and reaction rates. Note that the OSU database has been developed mainly for cold dark clouds. Therefore, references about BR for

Table 5. New HVC-SUBR for photo dissociation reactions.

Reactants	Total rate (10^{-9} s^{-1}) (OSU-01-2007)	Products	HVC-SUBR	OSU-01-2007-BR
$\text{C}_2\text{H} + h\nu$	1.0	$\text{C}_2 + \text{H}$	0.81	1.00
		$\text{C} + \text{CH}$	0.19	0.00
$\text{C}_3\text{H} + h\nu$	1.0	$\text{C}_3 + \text{H}$	0.65	1.00
		$\text{C}_2\text{H} + \text{C}$	0.33	0.00
		$\text{C}_2 + \text{CH}$	0.02	0.00
$\text{C}_4 + h\nu$	0.4	$\text{C}_3 + \text{C}$	0.77	0.50
		$\text{C}_2 + \text{C}_2$	0.23	0.50
$\text{C}_3\text{H}_2 + h\nu$	2.9	$\text{C}_3\text{H} + \text{H}$	0.46	0.66
		$\text{C}_3 + \text{H}_2$	0.23	0.34
		$\text{C}_2\text{H}_2 + \text{C}$	0.24	0.00
		$\text{C}_2\text{H} + \text{CH}$	0.04	0.00
		$\text{C}_2 + \text{CH}_2$	0.04	0.00
$\text{C}_4\text{H} + h\nu$	2.0	$\text{C}_4 + \text{H}$	0.58	0.50
		$\text{C}_3\text{H} + \text{C}$	0.26	0.00
		$\text{C}_3 + \text{CH}$	0.00	0.00
		$\text{C}_2\text{H} + \text{C}_2$	0.16	0.50
$\text{C}_5 + h\nu$	0.01	$\text{C}_4 + \text{C}$	0.13	0.00
		$\text{C}_3 + \text{C}_2$	0.87	1.00
$\text{C}_6 + h\nu$	1.0	$\text{C}_5 + \text{C}$	0.09	1.00
		$\text{C}_4 + \text{C}_2$	0.11	0.00
		$\text{C}_3 + \text{C}_3$	0.80	0.00
$\text{C}_7 + h\nu$	1.0	$\text{C}_6 + \text{C}$	0.01	1.00
		$\text{C}_5 + \text{C}_2$	0.19	0.00
		$\text{C}_4 + \text{C}_3$	0.80	0.00
$\text{C}_8 + h\nu$	1.0	$\text{C}_7 + \text{C}$	0.03	1.00
		$\text{C}_6 + \text{C}_2$	0.01	0.00
		$\text{C}_5 + \text{C}_3$	0.90	0.00
		$\text{C}_4 + \text{C}_4$	0.06	0.00
$\text{C}_9 + h\nu$	1.0	$\text{C}_8 + \text{C}$	0.00	1.00
		$\text{C}_7 + \text{C}_2$	0.06	0.00
		$\text{C}_6 + \text{C}_3$	0.66	0.00
		$\text{C}_5 + \text{C}_4$	0.28	0.00
$\text{C}_{10} + h\nu$	1.14	$\text{C}_9 + \text{C}$	0.01	0.82
		$\text{C}_8 + \text{C}_2$	0.01	0.17
		$\text{C}_7 + \text{C}_3$	0.70	0.00
		$\text{C}_6 + \text{C}_4$	0.03	0.01
		$\text{C}_5 + \text{C}_5$	0.25	0.00

Notes. Total rates and BR of OSU-01-2007 (<http://www.eric/o.html>) are also reported.

photodissociation processes are scarce. OSU-01-2007 Photodissociation BR for C_2H and C_3H used zero level statistical picture: only H emission is allowed. For C_4H , it assumed equal probability between emission of C_2 and H. For all C_nH species HVC-SUBR predict H emission to be by far the dominant channel. For C_3H_2 , OSU-01-2007 allows H or H_2 emission. Emission of C should also be an open channel. For the C_n series, as for the two previous processes, OSU-01-2007 assumed exclusive C emission. Exception arises from C_{10} where RRKM statistical calculations were performed (Bettens & Herbst 1995). Again HVC-SUBR predicts a stronger enhancement of the C_3 cluster production compared to OSU-01-2007.

4. Effects of the new branching ratios on chemical model predictions for dense clouds

4.1. Dense cloud model

In order to test the effect of the new branching ratios on chemical model predictions for dense clouds, we used the Nahoon

chemical model developed by V. Wakelam (Wakelam et al. 2004, <http://www.obs.u-bordeaux1.fr/amor/VWakelam>). This model follows the time-dependent chemistry of gas-phase species at a fixed temperature and density. We used Nahoon for a single spatial point (0D). We considered "typical" dense cloud conditions: a temperature of 10 K, an H density of $2 \times 10^4 \text{ cm}^{-3}$, a visual extinction (A_V) of 10 and a cosmic-ray ionization rate of $1.3 \times 10^{-17} \text{ s}^{-1}$. For the initial conditions, we started with all the elements but H in the atomic form with the low-metal elemental abundances from Graedel et al. (1982). The standard network used for this analysis is OSU-01-2007 (<http://www.physics.ohio-state.edu/~eric/research.html>), which contains 452 species and 4430 reactions. We updated the branching ratios listed in Tables 3–5 for 1) cosmic-ray dissociations of C_n ($n = 4$ to 9), C_nH ($n = 2$ to 4) and C_3H_2 , 2) dissociative recombinations of C_n^+ ($n = 5$ to 10), C_nH^+ ($n = 3$ to 4) and C_3H_2^+ , and 3) photo-dissociations of C_n ($n = 4$ to 10), C_nH ($n = 2$ to 4) and C_3H_2 . We then let the system evolve over 10^8 yr, when it reaches steady-state.

4.2. Results

Figure 10 shows the ratio between carbon-bearing species abundances computed with the updated database and those computed with the standard network OSU-01-2007. The new branching ratios modify the species abundances at two different times. The first one is a very early stage, before 10^4 yr, which is irrelevant for dense cloud chemistry. The second is much later, between 10^6 and 10^7 yr, but then the effect of the new branching ratios is less important. At the typical dense cloud age of 10^5 yr (Wakelam et al. 2006), the new branching ratios are unimportant. Higher ages have however been found for TMC-1 with other models (Terzieva & Herbst 1998), so the period around 10^6 yr is not without interest. After 10^6 yr, the maximum effect is obtained for the largest molecules. The C_nH molecules are an exception because C_7H and C_8H are more influenced than C_9H . All the C-bearing species abundances are decreased by the new BR by a maximum factor of two. The weak sensitivity of TMC-1 chemistry to BR has already been pointed out (Millar et al. 1988; Herbst & Lee 1997)

5. Effects of the new branching ratios on chemical model predictions for photon-dominated regions

5.1. PDR model

We tested the influence of these new rates on a PDR model. For this, we used the Meudon PDR code (<http://pdr.obspm.fr>) described in Le Petit et al. (2006). The Meudon PDR code computes the structure of a 1D plan-parallel and stationary slab of dust and gas. It consistently solves the radiative transfer from far UV to sub-millimeter, chemistry, and thermal balance. To test the influence of the new rates, we reproduced the model of the Horse Head by this PDR code presented in Pety et al. (2005) first with an old chemical network based on the rates provided by OSU-01-2007, secondly with the new branching ratios presented in this paper. The Horse Head is good candidate because of the large number of observed hydrocarbons. Pety et al. (2005) suggest that fragmentation of PAHs can contribute to the synthesis of small hydrocarbons and conclude by mentioning the need for precise chemical rates to perform chemical models in these regions. The proton density in the Horse Head is estimated to $n_{\text{H}} = 10^5 \text{ cm}^{-3}$ and the intensity of the incident UV flux to 100 times the ISRF (in Draine's units, Draine 1978). We adopted

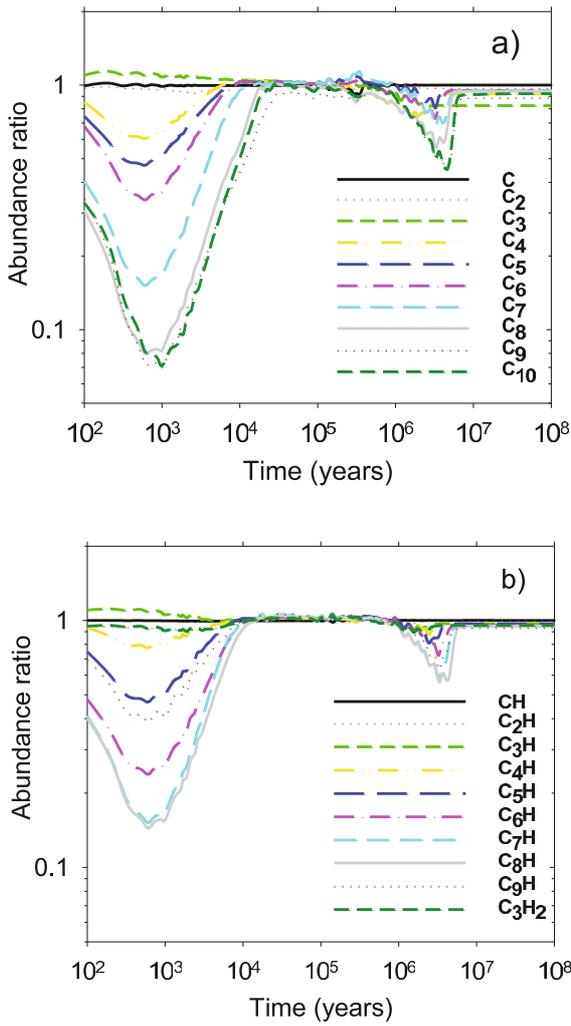


Fig. 10. Evolution with time of the ratios of abundances computed with the updated database and with the standard network (OSU-01-2007) for a dense clouds for a) C_n and b) C_nH_m species. (see text).

a flux of cosmic rays of 5×10^{-17} per H and per second. The model assumes a semi-infinite cloud. Our objective is to compare the profile of abundances of some relevant species computed with our new branching ratios and with older ones. To refine models of the Horse Head is beyond the scope of this paper.

5.2. Results

Figure 11 presents the ratios of abundances provided by the two chemistries of C_n and C_nH with n from 2 to 9 as a function of the position in the cloud expressed in visual extinction. This ratio is defined as the abundance provided with the new chemistry divided by the abundance obtained with a older rates. First we note that the effect of the new rates are only visible for A_V lower than 4. Indeed, it is in this region that the dissociative recombinations and photo-processes dominate the other chemical processes.

It is often difficult to reproduce the abundance of C_3 in PDR models (for example in the diffuse interstellar gas towards Zeta Persus, the model by [Le Petit et al. 2004](#), requires a high density component to reproduce this molecule). Figure 11 shows that the new branching ratios enhance the abundance of this molecule.

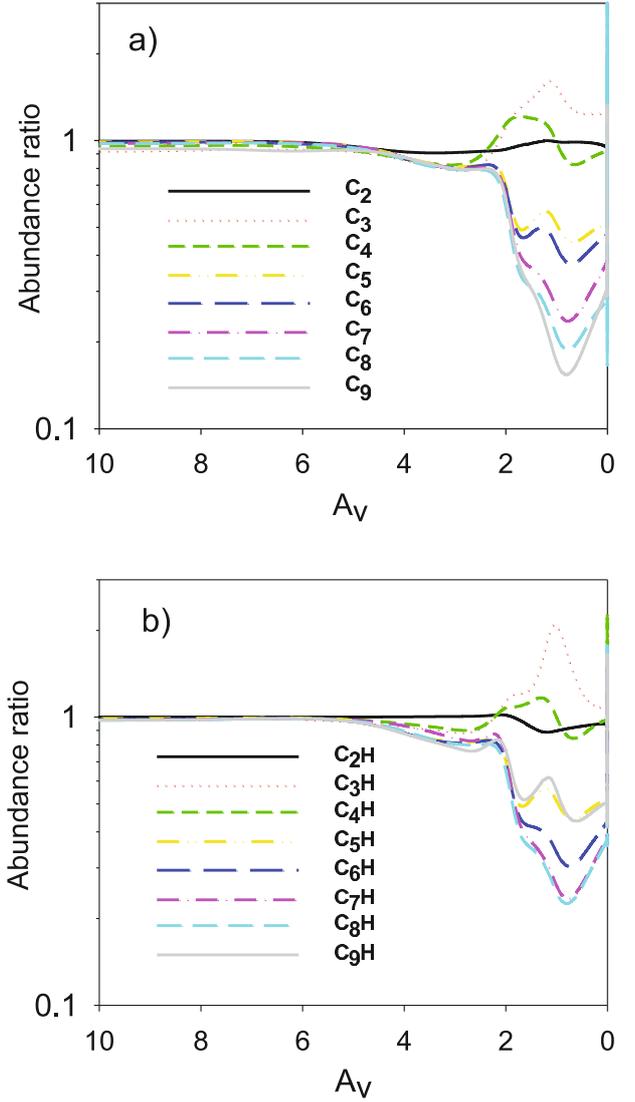


Fig. 11. Evolution, with the **visual** extinction A_V , of the ratios of abundances computed with the updated database and with the standard network (OSU-01-2007) for a PDR a) C_n and b) C_nH_m species (see text).

This is explained by two factors. First, the new branching ratios of the C_3H^+ recombination reaction enhance the route leading to C_3 by 30%. Secondly, we showed that the dissociative recombination of C_6^+ can efficiently produce two C_3 molecules. This route was not considered in previous chemistries.

For C_n molecules with $n > 4$, the new branching ratios systematically produce a decrease in the abundances. The model shows that the abundance of C_3H is significantly enhanced in the photodissociation area (Fig. 11b). This is explained by the new photodissociation route of C_4H . This route was not considered in the OSU database.

Finally, the abundances of the hydrocarbons, C_nH molecules ($n > 3$), are reduced compared to the old chemistry. This is directly linked to the decrease in the abundances the C_n molecules. Indeed, the chain of reactions leading to the hydrocarbons with $n > 3$ is



Because the abundance of C_n molecules is reduced by the new branching ratios, the abundances of the C_nH are also reduced.

6. Conclusions

High velocity collision in inverse kinematics scheme was used to measure the complete fragmentation pattern of electronically excited C_n ($n = 2$ to 10), C_nH ($n = 2$ to 4) and C_3H_2 molecules. Branching ratios of dissociation were deduced from those experiments. The comparison between the branching ratios obtained in this work and other types of experiments showed a good agreement. It was interpreted as the signature of a statistical behavior of the fragmentation. We thus propose new branching ratios for: 1) the dissociation of C_n ($n = 2-10$), C_nH ($n = 2-4$) and C_3H_2 molecules by interstellar UV photons or cosmic-ray processes and 2) the dissociative recombination of C_n^+ ($n = 5-10$), C_nH^+ ($n = 3-4$) and $C_3H_2^+$. The new values have been tested in dense cloud and PDR models. We showed that only chemistry occurring at A_V smaller than 4 is really affected. We however recommend astrochemists to use these branching ratios, even for dense chemistry, because it is well known that the importance of a reaction depends on the network we use. The data published in this paper have been added to the online database KIDA (Kinetic Database for Astrochemistry, <http://kida.obs.u-bordeaux1.fr>).

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