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Daniel Gautier
Observatoire de Paris

Berengere Dubrulle
French National Centre for Scientific Research

THE SOLAR SYSTEM D/H RATIO: OBSERVATIONS AND THEORIES

FRANÇOIS ROBERT[1], DANIEL GAUTIER[2], AND BÉRANGÈRE DUBRULLE[3]

[1]*Laboratoire de Minéralogie, CNRS-Muséum, Muséum d'Histoire Naturelle, 75005 Paris, France*
[2]*DESPA, URA 264, Observatoire de Meudon, F-92195, Meudon, France*
[3]*CNRS, UPR 182, Service d'Astrophysique, CES/Saclay, F-91191 Gif sur Yvette, France*

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Abstract. The measured D/H ratios in interstellar environments and in the solar system are reviewed. The two extreme D/H ratios in solar system water - $(720\pm120)\times10^{-6}$ in clay minerals and $(88\pm11)\times10^{-6}$ in chondrules, both from LL3 chondritic meteorites - are interpreted as the result of a progressive isotopic exchange in the solar nebula between deuterium-rich interstellar water and protosolar H_2. According to a turbulent model describing the evolution of the nebula (Drouart *et al.*, 1999), water in the solar system cannot be a product of thermal (neutral) reactions occurring in the solar nebula. Taking 720×10^{-6} as a face value for the isotopic composition of the interstellar water that predates the formation of the solar nebula, numerical simulations show that the water D/H ratio decreases via an isotopic exchange with H_2. During the course of this process, a D/H gradient was established in the nebula. This gradient was smoothed with time and the isotopic homogenization of the solar nebula was completed in 10^6 years, reaching a D/H ratio of 88×10^{-6}. In this model, cometary water should have also suffered a partial isotopic re-equilibration with H_2. The isotopic heterogeneity observed in chondrites result from the turbulent mixing of grains, condensed at different epochs and locations in the solar nebula. Recent isotopic determinations of water ice in cold interstellar clouds are in agreement with these chondritic data and their interpretation (Texeira *et al.*, 1999).

1. Introduction

The origin of organic compounds and water in the solar system has received an increasing interest in astronomy and cosmochemistry because it has been realized that Hydrogen-bearing molecules in comets, planets and chondritic meteorites show a systematic deuterium enrichment relative to the molecular hydrogen of the solar nebula (Robert *et al.*, 1979; Kolodny *et al.*, 1980; Robert and Epstein, 1982; McNaughton *et al.*, 1981, 1982; Kerridge, 1985; Kerridge *et al.*, 1987; Robert *et al.*, 1987; Deloule and Robert, 1995; Deloule *et al.*, 1998). Since there is no nuclear source for deuterium in the Universe (Epstein *et al.*, 1976; Galli *et al.*, 1995), such an isotopic enrichment must proceed via chemical reactions for which D reacts faster than H (H and D stands for hydrogen and deuterium, respectively). Because of the low abundances of the species formed by these reactions, the main reservoir of D in the Universe is H and H_2. Commonly speaking, some chemical reactions in the Universe must fractionate the two isotopes. Therefore, deciphering the origin of organic compounds and water in the solar system hinges on a correct identification of these reactions.

The isotopic exchange between water and molecular hydrogen is a classical example which can be used for the definition of the isotopic fractionation parameters

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(Geiss and Reeves, 1972; Lécluse *et al.*, 1996):

$$\mathrm{HD} + \mathrm{H_2O} \underset{k_r}{\overset{k_f}{\rightleftharpoons}} \mathrm{HDO} + \mathrm{H_2}, \tag{1}$$

The deuterium enrichment factor f in Eq. 1 is defined as:

$$f(\mathrm{H_2{-}H_2O}) = \frac{\frac{1}{2}\,[\mathrm{HDO}]\,/\,[\mathrm{H_2O}]}{\frac{1}{2}\,[\mathrm{HD}]\,/\,[\mathrm{H_2}]} = \frac{(\mathrm{D/H})_{\mathrm{H_2O}}}{(\mathrm{D/H})_{\mathrm{H_2}}}. \tag{2}$$

Equation 1 does not imply that the thermodynamical equilibrium is reached between the reactants. f is noted $f(\mathrm{H_2\text{-}H_2O})$ indicating that the isotopic exchange takes place between the most abundant species H_2O and H_2 (this notation will be used hereafter for all types of reactions). In this reaction, water is systematically enhanced in deuterium relative to molecular hydrogen; that is $f(\mathrm{H_2\text{-}H_2O}) > 1$. Under equilibrium conditions, f will be noted f_{equi} and stands for the forward to the reverse reaction rate ratio; that is $f_{\mathrm{equi}} = k_f/k_r$. f_{equi} depends exclusively on the temperature (Richet *et al.*, 1977). In the geochemical literature f_{equi} is noted $\alpha(T)$ and is referred to as “the isotopic fractionation factor”. Reaction 1 can be extended to all kind of H-bearing molecules (organic molecules, ions *etc.*) and yields (almost) always an enrichment in D in H-bearing molecules relative to H_2; that is $f(\mathrm{H_2\text{-}XH}) > 1$.

In space, isotopic exchange reactions could take place in three main different environments:

1. In the solar nebula via a thermal isotopic exchange between molecular hydrogen and H-bearing compounds (Geiss and Reeves, 1972; Lécluse and Robert, 1994)
2. In the dense interstellar medium at $T < 20$ K via isotopic exchange between ionized species and molecules (Brown and Millar, 1989; Brown and Rice, 1981; Watson, 1976; Willacy and Millar, 1998; Yung *et al.*, 1988; *cf.* References I.)
3. In denser interstellar clouds (the so called Hot-Cores), at intermediate temperatures ($T < 200$ K), via isotopic exchange between radicals (H or D) and neutral molecules (Rodgers and Millar, 1996; *cf.* References II.)

Calculations and/or experimental determinations of the f values for different molecules and for these three types of environments are available in the literature. The present paper is an attempt to compare these theoretical f values with solar system data in order to derive the possible relations between solar and interstellar molecules.

2. Distribution of the D/H Ratio in the Universe (Table I)

The deuterium is formed during the Big-Bang and since, is destroyed in stars. The formation of D in supernova shocks (Colgate, 1973), if possible, does not seem relevant in the Galactic context (Epstein *et al.*, 1976). Therefore the D/H ratio of the interstellar medium should have decreased with time by the dilution of D-free hydrogen injected in space by supernovae or stellar winds (Galli *et al.*, 1995). However, the exact amount of the overall decrease of D with time in the Galaxies, remains controversial (Tytler *et al.*, 1996; Burles and Tytler, 1998). Recent measurements with the Hubble Space Telescope have given an accurate determination of the local interstellar medium: Linsky *et al.* (1993) reports D/H ratios of $(16.5\pm1.8)\times10^{-6}$ and $(14.0\pm1)\times10^{-6}$ within 1 kpc of the Sun and McCullough (1992) reported 14 ratios averaging at 15×10^{-6}. The remarkable agreement between these two sets of data suggests that there is a single D/H ratio for the average interstellar medium. Note however, that Gry *et al.* (1983) or Vidal-Madjar *et al.* (1983) have argued that the D/H ratio may vary according to the line of sight and that the concept of a single ratio for the interstellar medium might be meaningless. In this text, $(D/H)_{H_2} = (16\pm1)\times10^{-6}$ is used as a reference ratio to calculate the different f values for the present day interstellar molecules (see Eq. 2 and Table I). The Standard Big-Bang Model of nucleosynthesis predicts the universal abundance of D which depends in practice on the baryon-to-photon ratio (Schramm, 1998). Although controversial, the initial D/H ratio of the Universe could be around 50×10^{-6} (Geiss and Gloeckler, 1998; see Table I). Subsequently, destruction of D in stars have caused the interstellar D/H ratio to decrease during these last 15 Gyr.

2.1. The Present Day Interstellar D/H Ratio in Water and in Organic Molecules

A large enrichment in D is observed in dense molecular clouds in all detectable C-bearing molecules (the so called organic molecules). In these clouds $T \sim 10$ K and H = 10^3 cm^{-3}. No reliable data exist on H_2O since its low density in the gas phase prevents its detection from the ground. A compilation of the isotopic composition of various interstellar organic molecules is reported in Fig. 1 in the form of histograms and their weighted means are reported in Table I. The measured large deuterium enhancement ($f>1000$) does not seem explainable by grain-surface chemistry or by gas phase reaction involving neutrals but presumably results from ion-molecule reactions (Watson, 1974; Brown and Millar, 1989; *cf.* References I.). Ions are formed in the gas by irradiation of the nearby stars whose UV's penetrates deep inside the cloud. At a first approximation, the enrichment in D reflects the H_2D^+/H_3^+ and the CH_2D^+ / CH_3^+ ratios which result from:

$$H_3^+ + HD \longleftrightarrow H_2D^+ + H_2 \tag{3}$$

$$CH_3^+ + HD \longleftrightarrow CH_2D^+ + H_2. \tag{4}$$

For these two reactions, $f_{\text{equi}}(H_3^+\text{-}H_2)=\exp(227/T)$ and $f_{\text{equi}}(CH_3^+\text{-}H_2)=\exp(370/T)$ respectively (Van Dishoeck, 1999). Under the form of H_2D^+ and CH_2D^+, D is transferred to the final detectable products via series of reactions such as:

$$H_2D^+ + CO \longleftrightarrow DCO^+ + H_2, \tag{5}$$

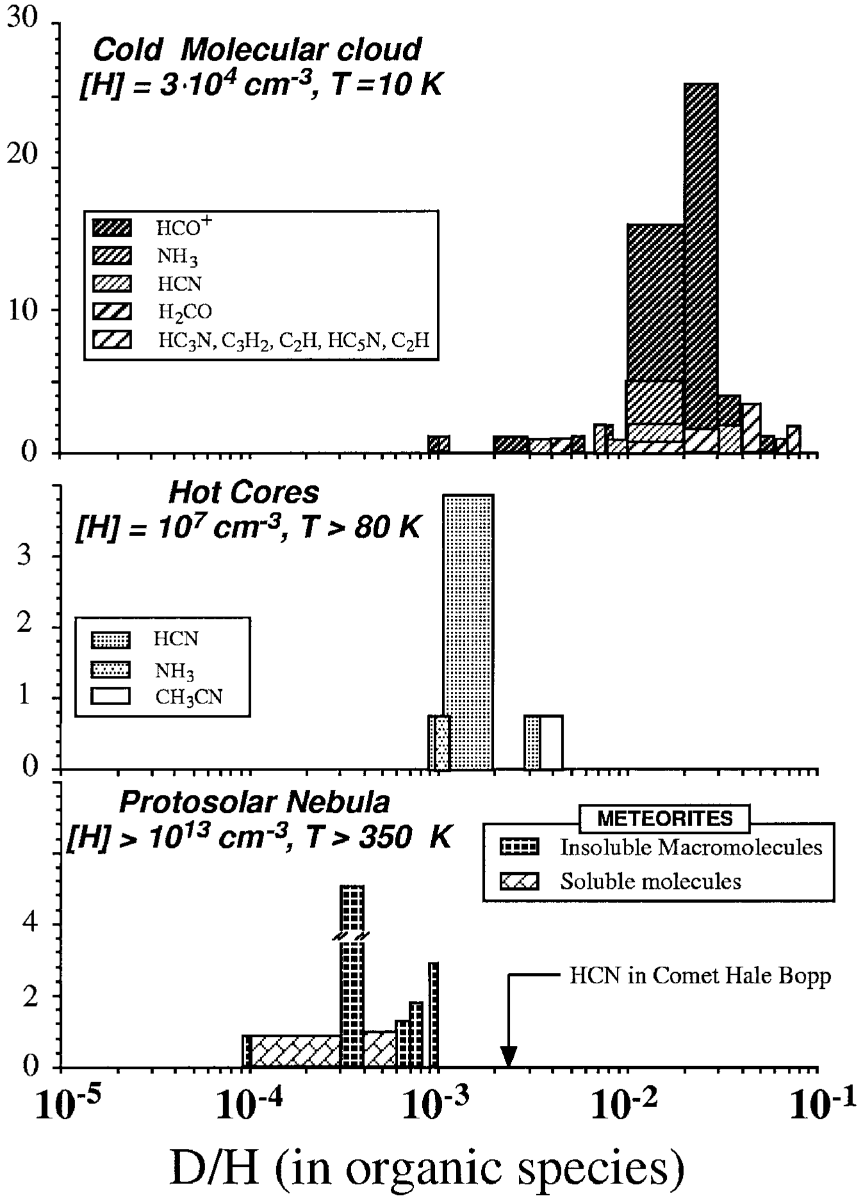

Figure 1. Histograms of distributions of the D/H ratio in organic molecules for three different environments (personal compilation of published data). These distributions are interpreted as follows: When temperature and density increase, the deuterated molecules formed at 10–20 K by ion-molecule reactions, become isotopically lighter and lighter via an isotopic exchange with H_2 (or H). Solar system organic compounds would also reflect such an isotopic processing occurring during the formation and evolution of the solar nebula.

TABLE I

Selected D/H ratios in Galactic and solar system objects expressed in values f=(D/H)$_{sample}$/(D/H)$_{ref}$. The reference value $f\equiv1$, representing the molecular hydrogen reservoir from which the H-bearing compounds are derived, has evolved with time. Therefore, f values are calculated for (D/H)$_{ref}$=16 or 25×10^{-6} for compounds formed recently or at the time when the solar system formed, respectively.

D/H (10^{-6})	**Species**	**Location**	**f**	**References**
Galactic[6]				
20 to 50	H	Big-Bang (Theoretical)	-	Schramm, 1998
34±2.5	H	Other Galaxy	-	Burles and Tytler, 1998
16±1	H	Local Interstellar Medium	≡1	Linsky *et al.*, 1993
110^{+150}_{-65}	H_2O	Hot Cores (HC)	7	Gensheimer *et al.*, 1996
1×10^{4} [10]	H_2O	Cold Interstellar Clouds (10 K)	625	Brown and Millar, 1989
$1^{+3}_{-0.2}\times10^{3}$	OM[1]	HC in Interstellar Clouds (IC)	63	Pers. compilation[3]
$2^{+0.7}_{-1.8}\times10^{4}$	OM[1]	Cold Interst. Clouds (<20 K)	1250	Pers. compilation[3]
950±550	H_2O (Ice)	Cold Interst. Clouds (10 K)	60±34	Texeira *et al.*, 1999
Proto Sun				
21±5	H_2	Nebula 4.5 Gyrs ago	≡1	Geiss and Gloeckler, 1998
Gaseous Planets[7]				
21±8	H_2	Jupiter	≡1	Lellouch *et al.*, 1996
26±7	H_2	Jupiter	≡1	Mahaffy *et al.*, 1998
15 to 35	H_2	Saturn	≡1	Griffin *et al.*, 1996
$65^{+2.5}_{-1.5}$	H_2	Neptune	2.6	Feuchtgruber *et al.*, 1999
55^{+35}_{-15}	H_2	Uranus	2.2	Feuchtgruber *et al.*, 1999
Comets[7]				
310±30	H_2O	Comet P/Halley	12.4	Eberhardt *et al.*, 1995
290±100	H_2O	Comet Hyakutake	11.6	Bockelée *et al.*, 1998
330±80	H_2O	Comet Hale-Bopp	13.2	Meier *et al.*, 1998a
2300±400	OM[1]	HCN in Comet Hale-Bopp	92	Meier *et al.*, 1998b
Interplanetary Particles[7]				
91–4018	(?)[11]	IDP and AMM[12]	3.6–161	Pers. compilation[3]
Telluric Planets[7]				
149±3	H_2O	Bulk Earth	6	Lécuyer *et al.*, 1998
16000±200	H_2O	Venus (*Atmosph. in situ*)	640	Donahue *et al*, 1982
780±80	H_2O	Mars (*Atmosph. in situ*)	31	Owen *et al.*, 1988
200–780	–OH	SNC (*Mars mantle*)	8–31	Watson *et al.*, 1994
LL3.0 and LL3.1 Meteorites[7]				
Interstellar compounds[8]				
730±120	–OH	in Clays and Chondrules	29	Deloule and Robert, 1995
800–1100	OM[1]	in Kerogen - like in Matrix	32–44	Pers. compilation[3]
Protosolar water[9]				
88±11	–OH	in Chondrules	3.5	Deloule *et al.*, 1998
Carbonaceous Meteorites[7]				
140±10	–OH	Mean Statistical Value	5.6	Pers. compilation[3]
130<D/H<170 (66%)[2]				
380–620	OM[1]	Kerogen - like in Matrix	15–25	Pers. compilation[3]
(CM,CV,CR)[4]				
370±6	OM[1]	Kerogen - like in Matrix	15–25	Halbout *et al.*, 1990
(Orgueil CI)[5]				
315–545	OM[1]	Amino Acids	12.5–22	Pizzarello *et al.*, 1991
185–310	OM[1]	Hydrocarbons and Carboxylic acids	7.5–12.5	Pizzarello *et al.*, 1991

Notes to Table I: (1) OM stands for Organic Matter. (2) Statistical Distribution. (3) Pers. compilation of published data (4) CM, CV, CR: Chondrite types. (5) The reference meteorite sample for the protosolar chemical composition. (6) $(D/H)_{ref}=16\times10^{-6}$. (7) $(D/H)_{ref}=25\times10^{-6}$. (8) The highest D/H ratios measured in LL3 are assumed to represent a minimum value for the interstellar water initially present in the solar nebula. (9) The lowest D/H value ("protosolar water") stands for interstellar water that underwent an isotopic exchange with the protosolar hydrogen; it is calculated from the statistical mean of D/H ratios measured in LL3 chondrites in the range $62–99\times10^{-6}$. (10) Theoretical values. (11) Not determined experimentally. (12) Interplanetary Dust and Antarctic Micro Meteorites.

$$CH_2D^+ + N \longleftrightarrow DCNH^+ + H_2 \qquad (6)$$

leading, in this example, to $f(HCO^+\text{-}H_2)$ and $f(HCN\text{-}H_2)$ = 1000 for $T < 50$ K (Guélin *et al.*, 1982; Duley and Williams, 1984). The steady state is reached in 10^7 years, a duration in agreement with the estimated life time for these cold media. Brown and Millar (1989) have shown that many other reactions are involved in the transfer of D from HD to ions (as for example, $C_2H_2^+$+HD). They have calculated that the measured distribution of D/H ratios corresponds to a duration close to 3×10^5 years. In recent and more detailed models, numerous additional reactions are now considered (Willacy and Millar, 1998) but it remains admitted that the deuterium chemistry in cold interstellar clouds is globally understood. Determinations of the D/H ratios in organic molecules and in water have been reported in molecular "Hot Cores". Hot Cores are warm (>100 K), dense ($H > 10^7$ cm^{-3}) and result – as in the case of Orion-KL - from the formation of young massive stars which heat the surrounding interstellar medium. Spatial resolution of the D/H ratio in HCN reveals the occurrence of a transition between the cold molecular cloud and the Hot Core region (Hatchell *et al.*, 1998; Schilke *et al.*, 1992): Systematically, in Hot Cores, the D/H ratios are lower than their cold interstellar counterpart (see Fig. 1). Detailed calculations show that the observed D enrichment cannot result from gas-phase chemistry: at Hot Cores temperatures (*i.e.* for $T > 70$ K), the f values should be essentially ≤ 10 *i.e.* no isotopic enrichment is expected for organic molecules or water (Millar *et al.*, 1989). Therefore, it has been proposed that, in Hot Cores, water results from the evaporation of icy mantles condensed at much lower temperature ($\sim$10 K) in the surrounding interstellar medium (Rodgers and Millar, 1996). In Hot Cores the composition of the gas is mainly neutral (Schilke *et al.*, 1992) and the deuterium exchange proceeds via reactions such as:

$$H + DCN \longleftrightarrow HCN + D. \qquad (7)$$

During this reaction f(HCN-H) decreases with time *i.e.* deuterium-rich HCN re-equilibrates with H at 150 K and is isotopically lighter than in the surrounding cold interstellar medium ($\sim$10 K). A similar situation seems to exist for H_2O: Measured D/H ratios in Hot Core H_2O vapor are systematically lower than those calculated for H_2O in cold molecular could. Therefore it has been proposed that, after its eva-

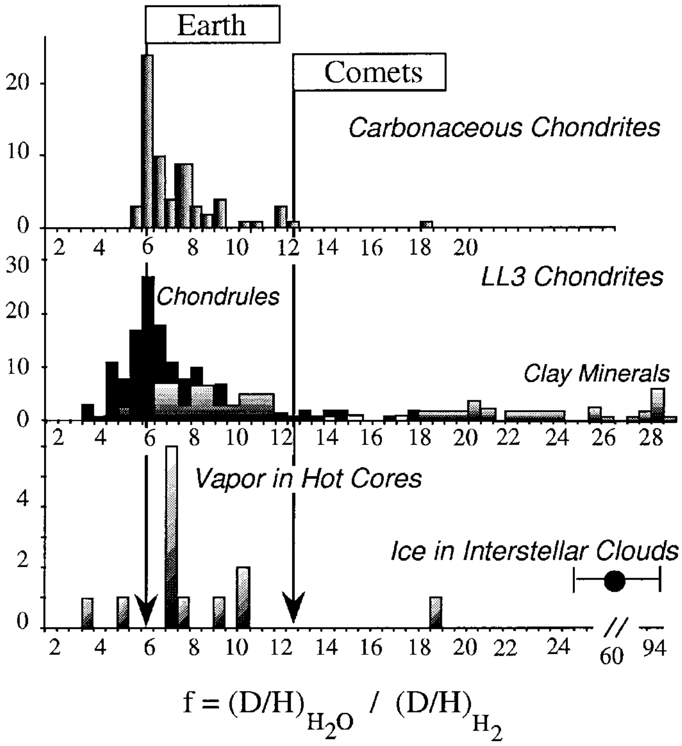

Figure 2. Histograms of distributions of the water D/H ratio in Carbonaceous Chondrites, in LL3 chondrules and clays, in Hot Cores and in interstellar ice (personal compilation of published data). According to this diagram, LL3 chondrites exhibit the best preserved record of the primordial isotopic heterogeneity of the solar system water. Note the similarity between the high D/H values in LL3 chondrites and in interstellar ices.

poration from the grains, water exchanges its D with atomic H, yielding a decreases in the D/H ratio while the temperature increases. As we will show in Sect. 3, the similarity between $f(H_2\text{-}H_2O)$ in Hot Cores and in meteorites (see Fig. 2) is an indication that isotopic exchange between water and H_2 can take place in space or in the solar nebula and yield a decrease of the D/H ratio in water. Note however, that these Hot Core regions cannot be taken as analogs of the presolar cloud from which the solar nebula formed. A recent estimate of the D/H ratio of solid water in interstellar mantle grains gives f=60±34.

2.2. The Early Sun and the Giant Planets

The D/H ratio in the solar nebula ($25\pm5\times10^{-6}$) is estimated from two independent determinations of (1) the Jovian and Saturnian D/H ratios and of (2) the present day solar $^3He/^4He$ and $^4He/H$ ratios.

1. Numerous spectroscopic determinations of the D/H ratio in the upper atmospheres of the two giant planets Jupiter and Saturn have been attempted (Beer and Taylor, 1973; 1978; Bezard *et al.*, 1986; de Bergh *et al.*, 1986; 1990; Feuchtgruber *et al.*, 1997; 1999; Griffin *et al.*, 1996; Lellouch *et al.*, 1996; Mahaffy *et al.*, 1998; Niemann *et al.*, 1996; Smith *et al.*, 1989a; 1989b; *cf.* References IV.). According to Gautier and Owen (1983) the D/H ratio of the two planets should reflect the value of the solar nebula. In these planets, D

is essentially in the form of HD whose abundance has been measured by the Infrared Space Observatory (Lellouch *et al.*, 1996). These observations are in good agreement with the HD/H_2 ratio measured in situ by the Galileo probe mass spectrometer (D/H = $26\pm7\times10^{-6}$; see Table I; Niemann *et al.*, 1996).

2. The deuterium in the early Sun has been converted in ^{3}He by the thermonuclear reaction: D + H $\longrightarrow$ ^{3}He. Therefore the D/H ratio of the early Sun can be derived from the present day $^3He/^4He$ measured in the solar wind ($^3He/^4He_{solar}$ $\approx$ $(3.8\pm0.5)\times10^{-4}$; Geiss and Gloeckler, 1998), provided the solar nebula $^3He/^4He$ ratio - noted hereafter $(^3He/^4He)_{sn}$ - of the Sun is known:
$$(D/H)_{earlySun} = (^3He/^4He_{solar} -^3 He/^4He_{sn}) \times (^4He/H)_{sn} \qquad (8)$$
The $(^3He/^4He)_{sn}$ estimated from chondrites or from Jupiter data is the most inaccurate parameter in Eq. 8. Using the recent Jovian estimates, a $(^3He/^4He)_{sn} \approx 1.5\times10^{-4}$ (Mahaffy *et al.*, 1998) corresponds to a $(D/H)_{earlySun} = (21\pm5)\times10^{-6}$ (Geiss and Gloeckler, 1998; with $(^4He/H)_{sn} \approx 10^{-1}$).
Combining the two determinations we adopt 25×10^{-6} for the solar nebula value. This ratio is therefore used here to calculate the f values reported in Table I for all solar system data (that is $(D/H)_{H_2} = 25\times10^{-6}$ in Eq. 2).
As noted previously, there is a slight difference between this solar ratio and the present day interstellar ratio (D/H = 16×10^{-6}). Therefore for an identical D/H ratio in the interstellar medium and in the solar system the calculated enrichment factors f are different.

2.3. Water-Rich Giant Planets

Uranus and Neptune have large icy cores. They exhibit D/H ratios significantly higher than in Jupiter and Saturn (Feuchtgruber *et al.*, 1997). An interpretation of this enrichment is based on formation models of these two planets (Pollack *et al.*, 1996) where the cores of the planets grew up by accretion of icy planetessimals having high (D/H) ratio (noted $(D/H)_{Planetesim}$). Assuming that in the interior, water from the planetesimals and molecular hydrogen from the gaseous envelope was isotopically equilibrated at high temperature ($f_{equi}(H_2\text{-}H_2O)$ =1) at least once during the life time of the planet, the $(D/H)_{Planetesim}$ ratios were calculated to be somewhat higher than the present day (D/H) ratio in H_2 (Lécluse *et al.*, 1996) but definitively lower than the cometary (D/H) ratios. However the isotopic equilibrium assumption may not apply if the planets were not fully convective.

2.4. Telluric Planets

Mars and Venus are mentioned here only for heuristic purposes. In fact, the photodissociation of water in their upper atmospheres yields the production of H and D, subsequently lost in space. Since H is lost much faster than D, the two atmospheres have been enriched in D during these last 4.5 Gyr by a Raleigh distillation process (Donahue *et al*, 1982; Owen *et al.*, 1988). However, the theory is too imprecise to derive accurately the primordial D/H ratio of these planets; therefore their present day D/H ratios are irrelevant to the questions raised in this paper.

The D/H ratio on Earth has been recently estimated accurately (Lécuyer *et al.*, 1998; D/H = $149 \pm 3 \times 10^{-6}$). Contrary to Mars and Venus, evaporation in space should not have caused any change in the D/H ratio of the oceans (within $\pm 1\%$) since the end of the planetary accretion. This has been shown theoretically (by modeling the flux of H escaping to space; Hunten and McElroy, 1974) and experimentally (by analyzing through geological times the terrestrial kerogen D/H ratios; Robert, 1989).

2.5. Chondrites

There are two types of primitive meteorites where high D/H ratio has been found: LL3 and carbonaceous chondrites (referred hereafter to as CCs). The insoluble organic matter (IOM) has been extracted chemically from these two type of rocks and has been found to be systematically enriched in D: $f(H_2\text{-IOM})$ = 15 to 25 in CCs (with most values around 15) and $f(H_2\text{-IOM})$ up to 44 in LL3. The possible contamination of IOM during the chemical procedure has been carefully evaluated and is negligible ($<$ 7%; Halbout *et al.*, 1990). When associated with clay minerals, IOM exhibits systematically D/H ratios higher than those in water extracted from clays. The exact nature of the chemical relations between clays and IOM are unknown and it is thus impossible to determine if clays acted as catalyses for the organic synthesis. In the case of the CC Murchison, the soluble organic fraction (amino-acids, fatty acids, hydrocarbons; see Table I) has been separated from the bulk sample by organic solvent procedures and gives $f(H_2\text{-SOM})$ = 12.5 to 22 (Epstein *et al.*, 1987; Pizzarello *et al.*, 1991).

After the formation of these two types of chondrites a late circulation of water occurring at (or near) the surface of their parent bodies is at the origin of the clay minerals (Bunch and Chang, 1980). An often quoted, terrestrial analogy for this mechanism is "hydrothermalism". The D/H ratio of these minerals can be used to determine the water D/H ratio because the isotopic fractionation between clay minerals and liquid water is negligibly small ($<$10%) at the scale of the isotopic variations in the solar system. The D/H ratios of these clay minerals has been determined by two methods: (1) by mass balance in CCs (*i.e.* by subtracting from whole rock analysis the measured IOM D/H ratio; *cf.* Robert and Epstein, 1982) or (2) by *in situ* measurements with the ion-microprobe (Deloule and Robert, 1995; Deloule *et al.*, 1998). The terrestrial contamination of meteorites by atmospheric water has been demonstrated to be negligible (Engrand *et al.*, 1999).

Up to 70% of the silicates in Chondrites are present as chondrules. They are droplets of silicates (diameter 50 to 1000 μm) quenched (from few seconds to 1 hour) in the solar nebula (Grossman *et al.*, 1997; Hewins, 1988; 1989, 1997; Scott and Taylor, 1983; 1994; Sears *et al.*, 1992; 1995; Wasson *et al.*, 1995). They may represent the first occurrence of silicates in the solar nebula, formed by the melting of sub-micron precursors, possibly of interstellar origin (Chaussidon and Robert, 1995). In LL3 chondrules, two sources of water have been identified : (1) water with a low D/H ratio ($f(H_2\text{-}H_2O)$ = 3.5). This water was involved in the late hydrothermalism and (2) water with a high D/H ratio [$f(H_2\text{-}H_2O) = 16$], located most commonly in pyroxenes ($MgSiO_3$) and which seems to have preserved its isotopic composition during the formation of chondrules.

In the matrix surrounding the chondrules, some clay minerals exhibits high D/H ratios [$f(H_2\text{-}H_2O) = 29$] which have also escaped – *i. e.* along with the pyroxenes in chondrules – the isotopic re-equilibration during the late hydrothermalism. In the matrix, when measured at random at a scale of 10μm with the ion-microprobe, isotopic compositions show a broad distribution (see Fig. 2), likely caused by the intimate mixture of deuterium-rich clay minerals and broken chondrule fragments. The mechanism for the incorporation and preservation of these high D/H ratios in chondrules or in the matrix remains puzzling.

The distribution of the $f(H_2\text{-}H_2O)$ in LL3 chondrules mimics that of the CC's but with more extreme end members (*cf.* Fig. 2). During hydrothermalism, an isotopic exchange between deuterium-rich organics and water may have yielded this narrow distribution of the D/H ratios in CCs. On the contrary, such an isotopic homogenization via hydrothermalism seems insignificant for LL3's. *In this respect, LL3 chondrites exhibit the best preserved record of the primordial isotopic heterogeneity of the solar nebula.*

Interplanetary dust particles have been collected in space or in Antarctic ice. They show extremely large variations in their D/H ratios [$f(H_2\text{-}H_2O = 3.6$ to 161] but the carrier(s) of this D-rich hydrogen has not yet been clearly identified (McKeegan *et al.*, 1985; Zinner *et al.*, 1983; Messenger, 1997; Engrand *et al.*, 1999). Whatever the mineralogical or molecular host of this deuterium-rich component, it has no chondritic counterpart. However, it should be noted that the lowest f value [$f(H_2\text{-}H_2O) = 3.6$] measured in these micrometeorites seems close to that measured in chondrites.

2.6. COMETS

The water D/H ratio has been reported for three different comets (see Table I) with a mean $f(H_2\text{-}H_2O)$ value = 12. Such a value is a factor of 2 higher than the mean chondritic value but a factor 2.5 lower than clay minerals in some LL3 chondrites. In HCN, $f(H_2\text{-}HCN) = 92$; such a high f value does not have a chondritic counterpart [the maximum $f(H_2\text{-}IOM) = 44$ in LL3].

It can be seen (*cf.* Fig. 2) that the weighted mean composition of CCs corresponds precisely to that of the Earth [$f(H_2\text{-}H_2O) =6$]. If CCs are taken as the carrier of water on Earth, a minimum f value can be assigned for the primitive Earth : $f \geq 4$ (that is the minimum f value of the CCs carbonaceous chondrite distribution). These remarks implies that (1) no major isotopic fractionation has occurred during the formation of the oceans and (2) less than 10% of cometary water has been added to the terrestrial oceans.

3. Spatio-Temporal Variations of the D/H Ratio in the Solar Nebula

Drouart *et al.* (1999) have shown that, in the solar nebula, the spatial and temporal distributions of the D/H ratio in minor species such as water, depend on temperature and pressure. They based their analysis on an analytical model proposed by (Dubrulle, 1992; Dubrulle and Valdettaro, 1992; Dubrulle, 1993; Dubrulle *et al.*, 1995) which describes the evolution of the solar nebula after the collapse of the presolar cloud. Three parameters dictate the evolution of the disk: (1) the coefficient of turbulent viscosity α, (2) the initial mass of the solar nebula M_{D}, and (3) its initial radius R_{D}.

(1): Shakura and Sunyaev (1973) have defined the phenomenological parameter α describing the efficiency of the turbulent viscosity. In such a turbulent solar nebula, the transfer rate of the angular momentum from the Sun outwards to Neptune is dictated by the value of α.

(2) and (3): M_{D} and R_{D} expressed in solar mass and astronomical units, respectively, are defined at a time $t(0)$ corresponding to an epoch where the initial molecular cloud has collapsed, forming the Sun at its center and the protoplanetary nebula around it. For $t > t(0)$, the accretion process takes place within the disk, resulting in its outward expansion (R increases) while most of the material flows inwards and gets accreted onto the proto-Sun, leading in turn to a decrease of M. The accretion rate is entirely defined by α, M_{D} and R_{D}. At time t and location R, all relevant physical quantities X can be written under the form:

$$X(R,t) = f(M_{\mathrm{D}}, R_{\mathrm{D}}, \alpha)\ . \tag{9}$$

X can be the temperature T, the pressure P or the surface density Σ. As the solar nebula evolves with time, or as R increases, T, P. and α decreases.

Contrary to Dubrulle (1993) who specified one set of values for the three parameters M_{D}, R_{D}, and α, Drouart *et al.* (1999) defined ranges for their possible variations: $10^{-10}<\alpha<10$, $5<R_{\mathrm{D}}<350.$, and $0.03<M_{\mathrm{D}}<0.3$. These initial values were then reduced using two types of constraints: (I) constraints from giant planets formation models and (II) constraints from measured D/H ratios in the LL3 chondritic meteorites.

(I) Four constraints are derived from giant planet formation models (Pollack *et al.*, 1996; see References V.): (i) The life time of the solar nebula is estimated from the formation model of Uranus ($t \leq 16\times10^{6}$ yrs). (ii) The water condensation temperature for the Jovian satellites (around 5 AU) must have been reached at $t<3\times10^{-6}$ yrs. (iii) The minimum initial gas surface density (Σ) of the solar nebula should have been >15 g cm^{-2} at 20 AU in order to form Uranus within 16×10^{-6} yrs (iv) The angular momentum must have been transported outwards to 30 AU (*i. e.* to Neptune) in $t \leq 10^{5}$ yrs.

(II) One constraint is derived from the lowest measured D/H ratio in chondritic water. That is: $f(H_2\text{-}H_2O)$ = 3.5 (*i. e.* D/H = 88×10^{-6}; *cf.* Semarkona LL3 chondrite) should be reached at $\approx$3 AU (the chondrite forming region) at any time during the life time of the solar nebula.

The constraints (I) allow us to restrict M_D, R_D, and α, in the domains 0.03–0.30 solar Mass, 5–230 AU and 10^{-4}–10^{0} respectively. The constraint (II) restrict further these domains: $8 < R_D < 28$ and $3 \times 10^{-3} < \alpha < 1$, for the initial M_D range ($0.03 < M_D < 0.3$).

The gas and the sub-micron grains experienced both inward and outward movements caused by : the solar accretion (inwards) and the turbulence (outwards) through which the angular momentum is transferred to the periphery of the disk. These opposite movements result in a efficient mixing between the internal and the external zones (Cassen, 1994). Therefore the D/H ratio in water depends on 2 terms : The isotope exchange term between H_2 and H_2O (Reaction 1) and the mixing term. This can be schematically expressed as:

$$f(\mathrm{H_2} - \mathrm{H_2O})(R, t) \ = \ \text{Isotopic Exchange} \ + \ \text{Turbulent Water Diffusion.} \quad (10)$$

The relations between the three parameters M_D, R_D, α, and $f(H_2\text{-}H_2O)(R,t)$ are presented qualitatively in the following.

The isotopic exchange term has been derived for an infinite reservoir of molecular H_2 relative to water (Lécluse and Robert, 1994) - that is the case of the solar nebula since $P(H_2O)$ / $P(H_2) \approx 10^{-4}$:

$$\frac{d\left[f(\mathrm{H_2} - \mathrm{H_2O})\right]}{dt} = P_{\mathrm{H_2}} k_{\mathrm{r}}(T)\left[f_{\mathrm{equi}}(T) - f\right] \quad (11)$$

with $k_r(T)$ the rate constant for the reverse isotopic exchange reaction between water and molecular H_2 (see Eq. 1 for the definition of k_r), P_{H_2} the total pressure of molecular H_2, $f_{equi}(T)$ the $f(H_2\text{-}H_2O)$ value under thermodynamical equilibrium. From Eq. 11 it can be seen that f depends upon P and T which in turn, vary according to M_D, R_D; hence the relation between f and M_D, R_D.

The diffusion term depends upon the pressure and the surface density. It is proportional to the parameter α expressed in terms of the "Prandtl number". The diffusion acts as a "stirring force": the higher α the higher the tendency of the solar nebula to be isotopically mixed. For example, for low α values ($\alpha < 10^{-6}$), the solar nebula is isotopically zoned: No exchange takes place at the periphery of the disk where the temperature is too low while the isotopic equilibrium is rapidly reached in the hot and inner zones of the solar nebula [$f_{equi}(H_2\text{-}H_2O) \approx f(H_2\text{-}H_2O) \approx 1$]. On the contrary, for high α values, the whole solar nebula have a tendency to be rapidly isotopically homogeneous. Therefore only intermediate α values can preserve an isotopic gradient in the solar nebula; hence the relation between $f(H_2\text{-}H_2O)(R,t)$ and α.

Eq. 10 is valid only for water vapor. When water condenses, the isotopic diffusion rate in ice is so slow that the solid cannot re-equilibrate with vapor and ice preserves indefinitely the isotopic composition of the vapor at the time of its condensation. Therefore the D/H ratio of the planetesimals formed in the solar nebula can be calculated through Eq. 10 at a temperature corresponding to the water condensation temperature.

Two different initial situations have been envisaged for the calculation of $f(H_2\text{-}H_2O)(R,t)$: (1) Water is formed at high temperature by the oxidation of molecular H_2 under thermodynamical conditions [in such a situation $f(H_2\text{-}H_2O)(t{=}0) = 1$], (2) interstellar water having a high f value is initially mixed with molecular H_2.

In situation (1), the calculated $f(H_2\text{-}H_2O)$ values never reach the chondritic value (f=3.5) for any value of the parameters M_D, R_D, and α compatible with type I constraints. This result demonstrates that *water in the solar system cannot be a product of thermal (neutral) reactions occurring in the solar nebula.*

In situation (2) one have to assume a realistic f value for the interstellar water at initial conditions. The highest reported D/H ratio in solar system water corresponds to f=29 (measured in the clay minerals from the Semarkona LL3 chondrite; see Table I). Since any isotopic exchange with H_2 would tend to lower this value, f=29 was assumed to represent the closest estimate for the interstellar water present in the solar nebula. Note that the recent determination of the D/H ratio in interstellar ice from cold clouds is in perfect agreement with such an estimate (Texeira *et al.*, 1999; f=60±34). The corresponding variations with time and space of the D/H ratio in water at the "snow-line" are reported in Fig. 3. The "snow-line" proceeds through time toward the center of the solar nebula that becomes globally colder and colder, while ice becomes isotopically lighter and lighter. This model reproduces two crucial observational facts: Water in the outer zones of the solar nebula have high D/H ratios ($f > 10$ as recorded by cometary water) and water in the inner zones (≈3 AU) exhibits an f value similar to that measured in LL3 chondrites (f=3.5). This model reproduces also the isotopic heterogeneity observed in chondritic bodies ($3.5 < f < 29$ in LL3 chondrites). Indeed, microscopic icy grains, condensed far away in the solar nebula (between 50 and 100 AU), can reach 3 AU in 10^6 years via their inward transport (Cuzzi *et al.*, 1993). These grains will never evaporate since they always remain in the ice field defined in the Fig. 3.

4. Alternative Models for the Origin of the Cometary D/H Ratio

The Drouart *et al.*'s model provides a scenario for the origin of comets consistent with their D/H ratios (see Fig. 3) and with other formation models (Weidenschilling, 1997): Comets were formed within 10^5 years in the solar nebula, at 30–40 AU, before being expelled to the Oort cloud. Although composed mainly of H_2O in their cores, the D/H ratios of Uranus and Neptune do not corresponds to those measured in comets (see Fig. 3). This suggests that the atmospheres of these planets were never fully convective and consequently that the isotopic exchange between their water cores and their hydrogen envelopes, never came to completion. Other possible explanations present several difficulties (see Drouart *et al.*, 1999).

The f values in cometary water (f=12) are often interpreted in the astronomical literature as an evidence that interstellar water was preserved in the outer part in the

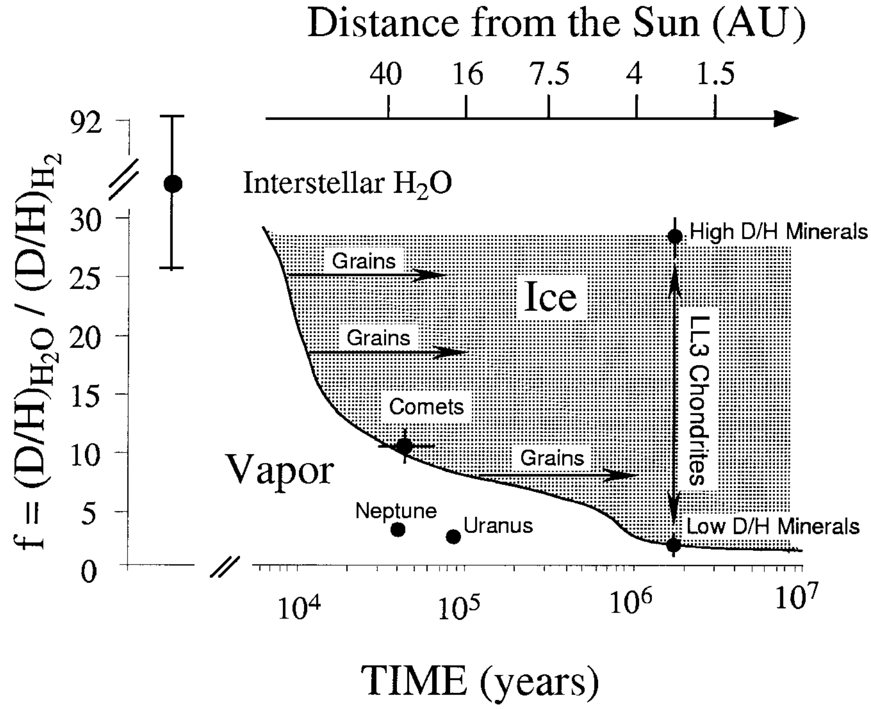

Figure 3. The D/H ratio of ice condensed in the solar nebula is reported as a function of time and space (Drouart *et al.*, 1999). The initial D/H ratio is 730×10^{-6} (*i. e.* f=29) as observed in clay minerals from the LL3 Semarkona chondrite and in interstellar ices. It can be observed that, since the solar nebula is colder and colder, the water condensation line (the snow-line) proceeds towards the center of the solar nebula through time. Large isotopic heterogeneities are created in the inner zones of the solar nebula for duration $>10^6$ years where icy grains are transported inward from distances up to 100 AU. As observed in LL3 chondrites, the water D/H ratio could vary between 730 and 85×10^{-6} at R <3 AU. One astronomical determination (Texeira *et al.*, 1999) of the D/H ratio in interstellar solid water is reported for comparison (400×10^{-6}<D/H<1500×10^{-6} *i. e.* f=60±34).

solar system (Meier *et al.*, 1998b). Such an interpretation, beside being in conflict with chondritic data, raised two difficulties briefly outline here after:

(1) According to Eq. 11, threshold values for temperature and pressure below which the isotopic exchange between water and hydrogen is kinetically inhibited, can be calculated. Below these values, the cometary water, even under the form of vapor, can preserve indefinitely its initial - interstellar - f value. That is $T\approx220$ K and $P\approx10^{-8}$ atm. Such $P - T$ conditions can certainly be fulfilled by other published models of the solar nebula but do not permit an interpretation of the D/H ratio in chondrites. Indeed, because of the turbulent nature of the solar nebula, the cometary forming zones must have been feed by water having f values $<<10$ and originated from the inner and hotter zones of the solar nebula. The present calculations show that such a dynamical addition of isotopically light water have a tendency to lower any interstellar f value, even for distances >30 AU. In other

words, an interstellar f value cannot be preserved in a turbulent solar nebula for reasonable values of the turbulent viscosity parameter ($\alpha > 10^{-4}$ or for plausible values of the Prandtl number; *cf.* Dubrulle, 1991). As a consequence, if an interstellar water D/H ratio was indeed preserved in the solar nebula, a mechanism that transfers the angular momentum without mixing the nebula, has to be found... Or comets did not form in the solar nebula...

(2) The second implication of having preserved interstellar D/H ratios in comets deals with the observation of chondritic f values (f=29) much higher than those in cometary water (f=12). In this scenario, the interstellar cloud that predates the formation of the solar nebula was isotopically heterogeneous and the interstellar water injected in the solar nebula at 3–4 AU had a f value of 29 but only of 12 in the regions of cometary formation. An explanation for this peculiar fact should be found. On the contrary, in the Drouart *et al.*'s model, large isotopic heterogeneities can be generated by the turbulence, in the inner solar system (see Fig. 3).

5. Conclusions

In conclusions we repeat the points we wish to emphasize. Water in the solar system cannot be a product of thermal (neutral) reactions occurring in the solar nebula. Water was initially synthesized by interstellar chemistry with a high D/H ratio $\geq 720\times10^{-6}$ (f=29). When in the solar nebula, its D/H ratio decreases via an isotopic exchange with H_2. During the course of this process, a gradient in D/H ratios was established in the solar nebula. This gradient was smoothed with time and the isotopic homogenization of the solar nebula was completed in no more than 10^6 years, reaching a D/H ratio of 88×10^{-6} (f=3.5). Cometary water should also have suffered a partial isotopic re-equilibration with H_2 before its condensation. The isotopic heterogeneity observed in chondrites result from the turbulent mixing of grains in the solar nebula, condensed at different epochs and locations.

6. Appendix: Compilation of References

6.1. References Quoted in the Text, Figures, and Table I

Beer and Taylor, 1973; Beer and Taylor, 1978; Bezard *et al.*, 1986; Bockelée *et al.*, 1998; Brown and Millar, 1989; Brown and Millar, 1989; Brown and Rice, 1981; Bunch and Chang, 1980; Burles and Tytler, 1998; Cassen, 1994; Colgate, 1973; Chaussidon and Robert, 1995; Chyba, 1987; Cuzzi *et al.*, 1993; de Bergh *et al.*, 1986; de Bergh *et al.*, 1990; Deloule and Robert, 1995; Deloule *et al.*, 1998; Van Dishoeck, 1999; Donahue *et al*, 1982; Drouart *et al.*, 1999; Dubrulle, 1991; Dubrulle, 1992; Dubrulle and Valdettaro, 1992; Dubrulle, 1993; Dubrulle *et al.*, 1995; Duley and Williams, 1984; Eberhardt *et al.*, 1995; Engrand *et al.*, 1999; Epstein *et al.*, 1976; Epstein *et al.*, 1987; Feuchtgruber *et al.*, 1997; Feuchtgruber *et al.*, 1999; Galli *et al.*, 1995; Gautier and Owen, 1983; Geiss and Reeves, 1972; Geiss and Gloeckler, 1998; Gensheimer *et al.*, 1996; Griffin *et al.*, 1996; Grossman *et al.*, 1997; Gry *et al.*, 1983; Guélin *et al.*, 1982; Halbout *et al.*, 1990; Hatchell *et al.*, 1998; Hewins, 1988; Hewins, 1989; Hewins, 1997; Hunten and McElroy, 1974; Kerridge, 1985; Kerridge *et al.*, 1987; Kolodny *et al.*, 1980; Lécluse and Robert, 1994; Lécluse *et al.*, 1996; Lécuyer *et al.*, 1998; Lellouch *et al.*, 1996; Linsky *et al.*, 1993; Lunine *et al.*, 1991; Mahaffy *et al.*, 1998; Messenger, 1997; McCullough, 1992; McKeegan *et al.*, 1985; McNaughton *et al.*, 1981; McNaughton *et al.*, 1982; Meier *et al.*, 1998a; Meier *et al.*, 1998b; Millar *et al.*, 1989; Niemann *et al.*, 1996; Owen, 1982; Owen *et al.*, 1988; Pizzarello *et al.*, 1991; Pollack *et al.*, 1996; Richet *et al.*, 1977; Robert, 1989; Robert and Epstein, 1982; Robert *et al.*, 1979; Robert *et al.*, 1987; Rodgers and Millar, 1996; Schilke *et al.*, 1992; Schramm, 1998; Scott and Taylor, 1983; Scott *et al.*, 1994; Scott and Taylor, 1983; Sears *et al.*, 1992; Sears *et al.*, 1995; Shakura and Sunyaev, 1973; Smith *et al.*, 1989a; Smith *et al.*, 1989b; Texeira *et al.*, 1999; Tytler *et al.*, 1996; Vidal-Madjar *et al.*, 1983; Wasson *et al.*, 1995; Watson, 1974; Watson, 1976; Watson *et al.*, 1994; Weidenschilling, 1997; Willacy and Millar, 1998; Yung *et al.*, 1988; Zinner *et al.*, 1983.

6.2. References I: D/H Ratios in the Dense Interstellar Medium – Theoretical Interpretations

Adams and Smith, 1987; Brown and Millar, 1989; Brown and Rice, 1981; Van Dishoeck, 1999; Duley and Williams, 1984; Geiss and Reeves, 1972; Henghman *et al.*, 1981; Millar *et al.*, 1989; Penzias, 1980; Schramm, 1998; Watson, 1974; Watson, 1976; Willacy and Millar, 1998; Yung *et al.*, 1988.

6.3. References II: D/H Ratio in Hot-Cores – Observations

Gensheimer *et al.*, 1996; Hatchell *et al.*, 1998; Jacq *et al.*, 1990; Rodgers and Millar, 1996; Schilke *et al.*, 1992; Walmsley *et al.*, 1987.

6.4. References III: D/H Ratio in the Dense Interstellar Medium – Observations

Bell *et al.*, 1988; Burles and Tytler, 1998; Charnley *et al.*, 1997; Combes *et al.*, 1985; Gottlieb *et al.*, 1979; Guélin *et al.*, 1982; Gry *et al.*, 1983; Herbst, 1982; Howe and Millar, 1993; Langer *et al.*, 1980; Linsky *et al.*, 1993; MacLeod *et al.*, 1981; McCullough, 1992; Olberg *et al.*, 1985; Penzias, 1979; Penzias *et al.*, 1977; Schloerb *et al.*, 1981; Turner, 1990; Vidal-Madjar *et al.*, 1983; Vrtilek *et al.*, 1985; Wilson and Rood, 1994; Wootten, 1987.

6.5. References IV: D/H Ratio in Solar System Objects – Observations

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6.6. References V: Chondrites and Chondrules – Observations

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6.7. References VI: Protosolar Nebula Models

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Address for Offprints: François Robert, Laboratoire de Minéralogie, CNRS-Muséum, Muséum d'Histoire Naturelle, 61 rue Buffon, 75005 Paris, France; `robert@cimrs1.mnhn.fr`