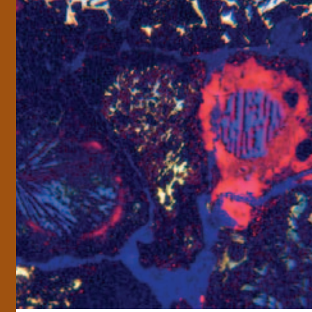

Organic Chemistry of Carbonaceous Meteorites

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The early Solar System contained a wide range of abiotic organic compounds. As the Solar System evolved, these organic molecules were incorporated into planetesimals and eventually planetary bodies, such as the parent bodies of meteorites. One particular class of meteorites, the carbonaceous meteorites, contains a large variety of extraterrestrial organic compounds. These compounds represent a record of the chemical reactions and conditions in the early Solar System. Different formation mechanisms and sources (interstellar, nebular or parent body) contributed to the inventory of meteoritic organic molecules. Their subsequent delivery to the early Earth may have contributed the first prebiotic building blocks of life.

Keywords: carbonaceous meteorites, soluble organic matter, extraterrestrial organic molecules, building blocks of life

INTRODUCTION

Some 20–30% of the mass of our Galaxy is in the form of gas and dust between the stars. This interstellar medium (ISM) consists primarily of gaseous atomic and molecular hydrogen (H) and helium (He), which account for ~99% of the total mass. The next most abundant atoms, oxygen (O), carbon (C), and nitrogen (N), collectively account for about 1% of the ISM's mass. The remaining elements are present only in trace amounts. Astronomical observations of the ISM reveal numerous organic compounds. More than 150 different chemical species, many of which are relatively complex, have been identified in interstellar and circumstellar regions. These chemical species provide the precursor material for planetary systems. A key question in origin-of-life research is how much of this material survives the brief but violent periods of cloud collapse, disk formation, and planetary accretion.

Meteorites are extraterrestrial objects that survive the passage through the Earth's atmosphere and impact the Earth's surface. The more primitive, less altered meteorites are called chondrites. Studies of carbonaceous chondrites provide evidence that interstellar organic matter survived planet-formation processes and was incorporated into asteroid-sized objects.

CI carbonaceous chondrites – a particular group of chondrites – have a chemical composition that matches the chemistry of the Sun more closely than any other class of meteorites (Anders and Grevesse 1989). Therefore, they are the most chemically primitive meteorites known. Other chondrite groups, particularly the CM and CR chondrites, show only slight depletions in volatile elements relative to the CI chondrites.

The CI, CM, and CR chondrites contain carbon in different forms, including silicon carbide, graphite, diamonds, carbonate and organic matter. These chondrites contain up to 2 wt% organic carbon, more than 70% of which is composed of a solvent-insoluble macromolecule (Cody et al. 2002; Cody and Alexander 2005), while less than 30% is a mixture of solvent-soluble organic compounds (for reviews see, for example, Botta and Bada 2002 and Sephton 2002). Study of these samples provides insight into the distribution of organic reservoirs in the Solar System and the specific organic compounds present in these reservoirs. Some of the organic compounds present in meteorites are important in terrestrial biochemistry. In particular, the amino acids are monomers of proteins, and the nucleobases are the informational monomers of RNA and DNA and have a prominent role in coenzymes. Carbonaceous meteorites exogenously delivered these important prebiotic organic molecules to the early Earth, leading some researchers to suggest that they contributed the first prebiotic building blocks of life (Chyba and Sagan 1992). They therefore provide information about the origin of life on Earth and the possible presence of life elsewhere in the Solar System.

Chondrites are grouped into petrographic types 1 to 6. This classification reflects the intensity of thermal metamorphism (i.e. the adjustment of the minerals on the meteorite parent body in response to increased temperature). It also reflects the intensity of aqueous alteration (i.e. the transformation of the original minerals into a new assemblage of minerals caused by low-temperature reaction with water) that occurred on the meteorite parent body. Petrologic types 2 and 1 indicate increasingly high aqueous alteration. Petrologic types 4 to 6 indicate increasingly high thermal metamorphism. Type-3 chondrites are minimally altered and are mineralogically and texturally the most primitive. The CI, CM, and CR chondrites are primarily of petrologic types 1 and 2, though a few CR chondrites are type 3. Thus, even though the CI, CM, and CR chondrites are chemically primitive, they have experienced a range of aqueous or hydrothermal alteration on their parent asteroid. Within the asteroidal parent bodies of these chondrites, the presumably simpler interstellar organic compounds were transformed into more complex organic compounds by aqueous and thermal processing.

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KEY METEORITE SAMPLES IN ORGANIC COSMOCHEMISTRY

The CI chondrites are samples of some of the most volatile-rich small bodies in the Solar System. Only seven members of this extraordinarily important meteorite group have been recovered, with a total mass of ~15.5 kg. Almost 90% of this material is represented by the Orgueil meteorite, which fell in May 1864 in the Midi-Pyrénées, France. Orgueil is the most volatile-rich meteorite known and contains over 3% carbon by weight. It serves as a chemical standard against which the compositions of all other meteorites are compared.

In September 1969, over 100 kg of a particularly important carbonaceous chondrite fell near Murchison, Victoria, Australia. The Murchison CM2 meteorite is rich in volatile compounds, containing over 2% carbon by weight. Due to its pristine nature and the fact that a large amount of the Murchison meteorite is available, its organic content has been extensively analysed. As a result, it serves as a baseline reference for all other carbonaceous chondrites analysed for organic compounds. Other important CM2 chondrites that have been widely analysed include Nogoya, Cold Bokkeveld, and Murray.

Tagish Lake is an ungrouped type-2 carbonaceous chondrite that fell in British Columbia, Canada, in January 2000. Following the reported sighting of a fireball in southern Yukon and northern British Columbia, more than 500 fragments of the meteorite, amounting to over 10 kg, were collected from the frozen surface of Tagish Lake. It is an important and unique extraterrestrial sample because it was recovered frozen, preserving many of its original organic features.

The US-led Antarctic Search for Meteorites (ANSMET) group—a collaborative effort by three government agencies (the National Science Foundation, the National Aeronautics and Space Administration, and the Smithsonian Institution)—and similar programs from other nations have collected an enormous number of invaluable meteorite specimens. Antarctic meteorites have been a scientific bonanza. Collection efforts over the past 30 years have recovered over 25,000 meteorite samples, including many unique, organic-rich carbonaceous chondrite samples. The name of an Antarctic meteorite consists of the complete geographical name together with the numerical code, e.g. LaPaz Icefield 03784 or Elephant Moraine 92042, where the first two digits correspond to the year in which the meteorite was found and the remaining digits correspond to the specimen number. Furthermore, the geographical locations are usually abbreviated, giving LAP 03784 and EET 92042 for the two meteorites above.

THE ORIGIN OF EXTRATERRESTRIAL ORGANIC MATTER

The first objective in the study of extraterrestrial organic molecules is to determine whether or not they are indigenous to the meteorites. Extraterrestrial organic molecules present in carbonaceous chondrites exhibit large structural diversity, which is a feature contrasting with terrestrial biogenic organic compounds. In addition, life is known to encode only 20 protein α-amino acids. Only eight of these terrestrial, biological amino acids have been identified in meteorite extracts. However, there are two strong lines of geochemical evidence for their extraterrestrial origin: the enantiomeric ratios in chiral molecules (i.e. molecules that have a non-superimposable mirror image) and the stable isotope values for hydrogen, carbon, and nitrogen.

Chirality is a useful tool for determining the origin of a compound present in meteorites. Pairs of enantiomers, i.e. the mirror images of a chiral molecule, are designated as either right-handed (D-) or left-handed (L-). On Earth, proteins and enzymes are made of only the L-enantiomer of chiral amino acids. On the other hand, the abiotic synthesis of amino acids yields racemic mixtures [i.e. with a ratio of the D- and L-enantiomers close to unity (D/L ~1)]. Racemic mixtures have been observed for most non-protein chiral amino acids (for a review see Martins and Sephton 2009). However, small L-enantiomeric excesses are reported for some non-protein amino acids in meteorites (Pizzarello et al. 2003; Glavin and Dworkin 2009). Glavin and Dworkin (2009) found the largest L-enantiomeric excess (Lee) of isovaline in any meteorite reported to date in the CM meteorite Murchison (Lee = 18.5%). The CI meteorite Orgueil also contained a similar enrichment in L-isovaline (Lee = 15.2%). This large asymmetry in the CI and CM meteorites suggests that amino acids delivered by

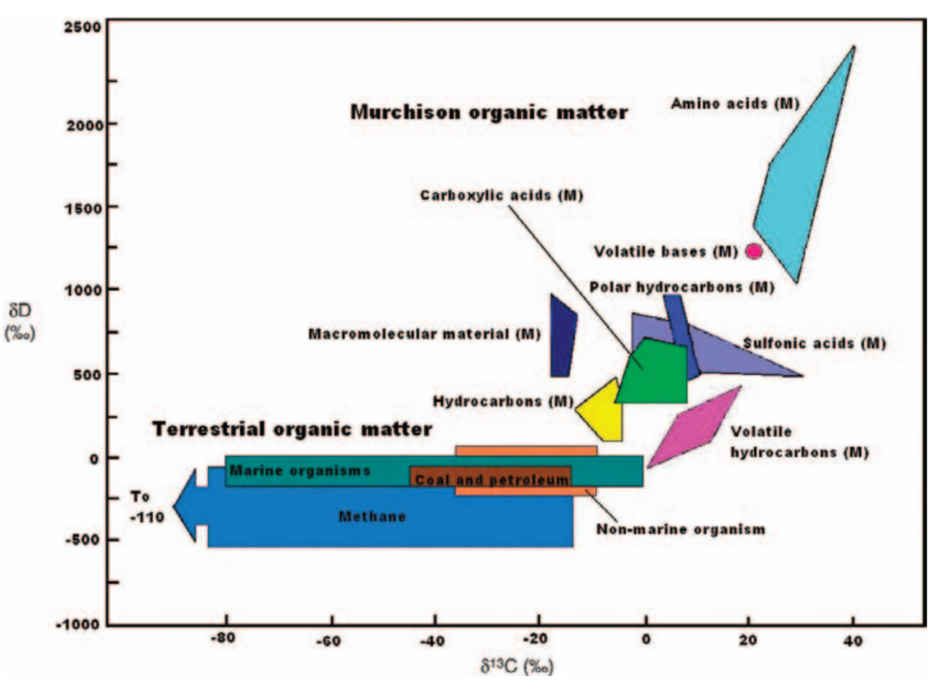

FIGURE 1 Stable carbon and hydrogen isotope ratio values in the Murchison meteorite and in terrestrial environments. Murchison organic matter has the symbol (M) after each organic compound. ADAPTED FROM SEPHTON AND BOTTA (2005)

asteroids, comets, and their fragments may have biased the Earth's prebiotic organic inventory with left-handed molecules before the origin of life.

For non-chiral compounds, stable isotope measurements are the only means to establish their origin (e.g. Cronin et al. 1993). The relative abundances of the heavy stable isotopes of carbon and hydrogen (^{13}C and D, respectively) in these compounds differ significantly from those in terrestrial materials. Organic materials formed in terrestrial environments (atmospheric, biomass-burning, methanogenic and thermogenic) have δD values ranging from –300‰ to –50‰, and δ^{13}C values ranging from –60‰ to –25‰ (Wahlen 1994). On the other hand, meteoritic organic compounds are substantially enriched in D, ^{13}C and ^{15}N, providing decisive evidence that these materials are not terrestrial contaminants (Fig. 1; e.g. Huang et al. 2005). Stable isotope measurements are thus critical for establishing the origin of organic components present in carbonaceous chondrites.

THE NATURE OF EXTRATERRESTRIAL SOLUBLE ORGANIC MATTER

Carbonaceous chondrites contain organic carbon in the form of a solvent-insoluble macromolecule (more than 70%) and a mixture of solvent-soluble organic compounds (less than 30%). The insoluble organic matter (IOM) can be isolated after solvent extraction (to remove soluble organic compounds), and treatment with hydrofluoric acid (to remove silicates) and hydrochloric acid (to remove carbonates). Analyses showed that the IOM consisted mainly of small aromatic molecules (for reviews see e.g. Sephton 2002). A large amount of research has, therefore, focused on the solvent-soluble fractions of carbon-rich chondrites; this research has been facilitated by the relative ease with which these compounds can be separated from the bulk sample (typically a few hundred milligrams of sample) using solvents. Some carbonaceous chondrites contain a spectacularly complex suite of soluble organic molecules (Table 1). These include amino acids, carboxylic acids, purines and pyrimidines, and hydrocarbons, among others (see Figure 2 for the chemical structure of these molecules; for a review see Botta and Bada 2002, Sephton 2002, and Pizzarello et al. 2006). Concentrations of the major representatives of these classes vary widely: less than 10 parts per million for amines, tens of parts per million for amino acids, and hundreds of parts per million for carboxylic acids. Here, I review the soluble organic fraction of the organic carbon present in carbonaceous chondrites, focusing on new results from research on amino acids, carboxylic acids, nucleobases (purines and pyrimidines), and polycyclic aromatic hydrocarbons (PAHs).

Amino Acids

Amino acids are an important component of the soluble fraction of meteorites and have attracted a lot of attention because of their central role in modern biochemistry. Their distribution varies greatly among different carbonaceous chondrites (Martins and Sephton 2009). More than 80 different amino acids have been identified in the Murchison meteorite (Martins and Sephton 2009). Most of these amino acids, like α-aminoisobutyric acid (α-AIB) and isovaline, are rare in the terrestrial biosphere and are not protein-forming amino acids.

While Murchison (CM2) contains amino acids showing large structural diversity (the most abundant structural type is α, followed by β and γ configurations), Orgueil (CI1) has much less diversity, with β-alanine as the most abundant amino acid. There is also considerable variation in

Table 1 ABUNDANCES OF SOLUBLE ORGANIC MATTER IN THE MURCHISON AND TAGISH LAKE METEORITES

	Abundances (ppm)	
Compounds	**Murchison**	**Tagish Lake**
Carboxylic acids (monocarboxylic)	332	40
Sulfonic acids	67	≥20
Amino acids	60	<6
Dicarboximides	>50	5.5
Dicarboxylic acids	>30	17.5
Ketones	17	n.d.
Hydrocarbons (aromatic)	15–28	≥1
Hydroxycarboxylic acids	15	b.d.
Hydrocarbons (aliphatic)	12–35	5
Alcohols	11	n.d.
Aldehydes	11	n.d.
Amines	8	<0.1
Pyridine carboxylic acid	>7	7.5
Phosphonic acid	1.5	n.d.
Purines	1.2	n.d.
Diamino acids	0.4	n.d.
Benzothiophenes	0.3	n.d.
Pyrimidines	0.06	n.d.
Basic N-heterocycles	0.05–0.5	n.d.

b.d. – below the detection limit; n.d. – not determined

Adapted from Pizzarello et al. 2001, Botta and Bada 2002, Sephton 2002, and Glavin et al. 2010

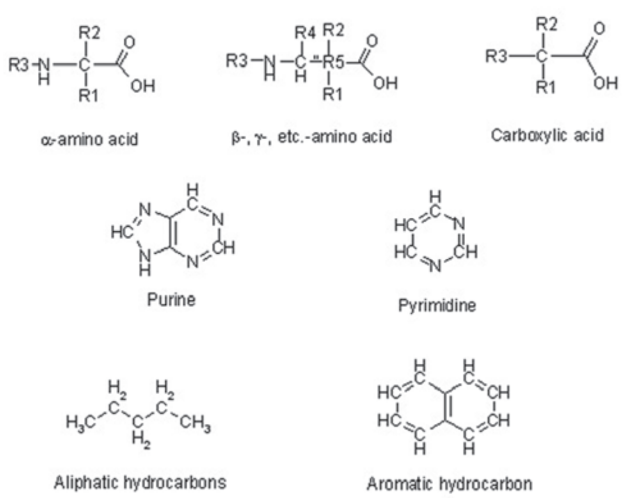

Figure 2 Chemical structure of amino acids (α-, β-, γ-, etc.), carboxylic acids, nucleobases (purines, which are two-ring N-containing heterocyclic compounds, and pyrimidines, which are one-ring N-containing heterocyclic compounds), and hydrocarbons (aliphatic and aromatic). R1, R2, R3 and R4 correspond to a hydrogen or $C_nH_{(2n+1)}$. R5 corresponds to an alkyl group. The carbon next to the carboxyl group (-COOH) is called α-carbon, and the carbons in the carbon chain are labelled α-, β-, γ-, δ-, and so on. The amino acids take their name from the position the amine group (-NH_2) is attached to the carbon chain, i.e. an amino acid with the amine group attached to the α-carbon is called α-amino acid.

the abundance of amino acids among other carbonaceous chondrites. The Antarctic CR2 meteorites Elephant Moraine (EET) 92042 and Graves Nunatak (GRA) 95229 (Fig. 3) have the highest known abundances of amino acids, with total amino acid concentrations of 180 parts per million (ppm) and 249 ppm, respectively (Martins et al. 2007). The extraterrestrial origin of these amino acids is indicated by racemic enantiomeric ratios (D/L ~1) and high values of $\delta^{13}C$ (Martins et al. 2007), well outside the carbon isotope range of terrestrial amino acids (Scott et al. 2006). Other indications of an extraterrestrial origin include D and ^{15}N enrichments (Pizzarello et al. 2008; Pizzarello and Holmes 2009), which are interpreted as a result of chemical fractionation in the interstellar medium. In addition, the CR2 La Paz (LAP) 02342 (Pizzarello and Holmes 2009) and the CR1 Grosvenor Mountains (GRO) 95577 have been analysed for amino acids. The latter is depleted in amino acids, containing just 1 part per billion (ppb) (Martins et al. 2007). Among the CM chondrites there is also heterogeneity in the amino acid abundance, with some essentially free of amino acids, such as the CM1 chondrites ALH 88045, Meteorite Hills (MET) 01070 and LAP 0227 (Botta et al. 2007).

The formation of meteoritic amino acids likely proceeded through different synthetic pathways for different structural types. α-amino acids were likely synthesised in a two-step process in which α-amino acid precursors (i.e. carbonyl compounds, ammonia and hydrogen cyanide) formed in the protosolar nebula and were later incorporated into an asteroid; subsequent aqueous alteration in the asteroid induced Strecker-cyanohydrin synthesis and the production of α-amino acids (Fig. 4). On the other hand, meteoritic β-amino acids may have formed via Michael addition of ammonia to α,β-unsaturated nitriles, followed by reduction/hydrolysis (Fig. 5). In addition, the hydrolysis of lactams gives the corresponding β-, γ- and δ-amino acids (Cronin and Chang 1993).

When analysing potential meteoritic parent bodies, it is interesting to compare the organic inventory in carbonaceous chondrites with that in comets. Carbon isotope measurements of amino acids present in cometary samples returned to Earth by NASA's Stardust spacecraft revealed a $\delta^{13}C$ value for glycine of +29‰ (Elsila et al. 2009). This value falls in the range previously reported for glycine in the Murchison meteorite ($\delta^{13}C$ = +22‰ to +41‰) and the CI-type meteorite Orgueil ($\delta^{13}C$ = +22‰) (Pizzarello et al. 2006; Martins and Sephton 2009). These results further strengthen the connection between cometary samples and primitive chondrites (see Gounelle 2011 this issue).

Carboxylic Acids

Carboxylic acids are the most abundant class of soluble compounds in Murchison. The Murchison and Murray (CM2) meteorites contain monocarboxylic acids ranging from C_2 to C_{10}, with abundances ranging from 10 to 60 ppm for each compound (Huang et al. 2005). Carbon isotope ratios range from –31.8‰ to +32.5‰. All species of these acids have positive values of δD, except for the C9 and C10 straight-chain monocarboxylic acids. Straight-chain monocarboxylic acids show a relationship between increasing chain length and enrichment in lighter isotopes, which supports a kinetically controlled synthesis involving radical-induced carbon addition reactions. Pizzarello et al. (2001) reported total concentrations of monocarboxylic acids around 40 ppm in Tagish Lake and >300 ppm in Murchison (Table 1). Monocarboxylic acids have also been reported in the Antarctic carbonaceous chondrites EET 96029 (Huang et al. 2005), Yamato (Y-) 74662, Asuka (A-) 881458, A-88120 and A-881334 (Shimoyama et al. 1989; Naraoka et al. 1999), with monocarboxylic acids ranging from C2 to C12.

The Tagish Lake meteorite also contains dicarboxylic acids with a total concentration of 17.5 ppm, along with aromatic dicarboxylic acids as a series of imide derivatives of phthalic acid, homophthalic acid, and some alkyl homologues, in benzene/methanol extracts (Pizzarello et al. 2001). Dicarboxylic acids from the Tagish Lake sample have $\delta^{13}C$

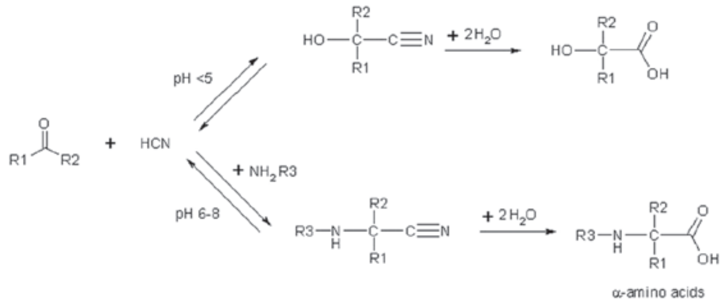

Figure 4 Strecker synthesis for the formation of α-amino acids (adapted from Botta and Bada 2002). R1 and R2 correspond to a hydrogen or $C_nH_{(2n+1)}$. If R3 corresponds to hydrogen, then α-amino acids are produced. When hydrogen cyanide and aldehydes are present in the parent body of carbonaceous chondrites, and if the pH is acidic (pH <5) and the abundance of ammonia is low (or there is no ammonia), then α-hydroxycarboxylic acids are produced. On the other hand, if the pH is higher (pH 6 to 8) and ammonia is present, then α-amino acids are formed.

Figure 3 GRA 95229 meteorite sample after preparation by the Antarctic meteorite curator at NASA Johnson Space Center

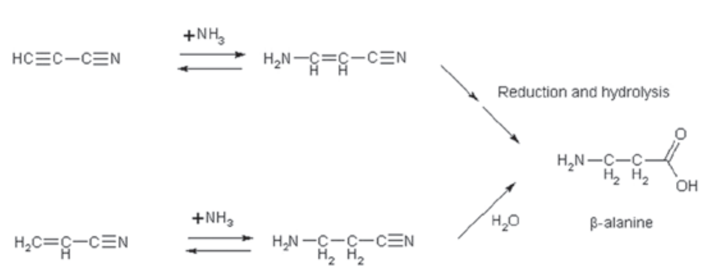

Figure 5 Synthesis of β-amino acids through Michael addition of ammonia to α,β-unsaturated nitriles, followed by reduction and hydrolysis (adapted from Botta and Bada 2002)

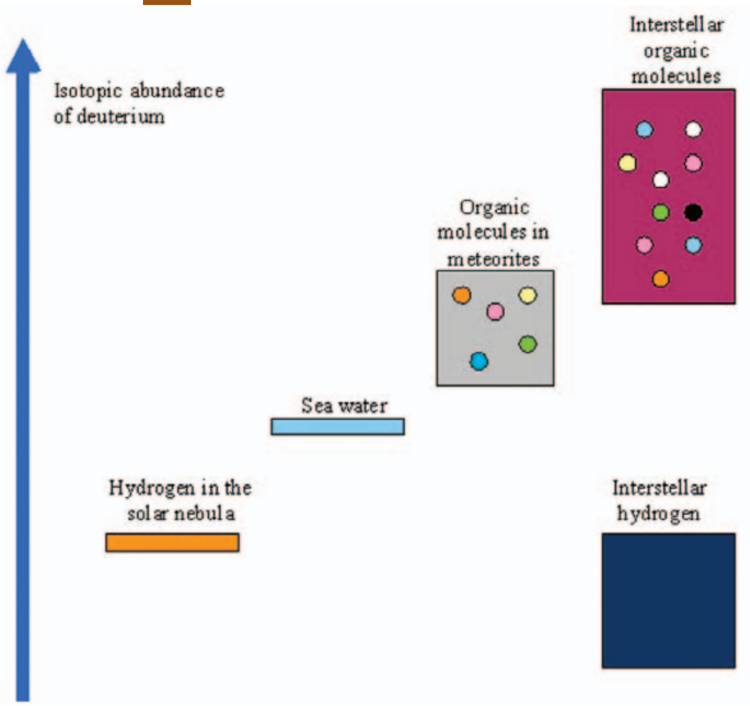

FIGURE 6 Abundance of deuterium in diverse regions of the Universe. Interstellar hydrogen and hydrogen in the solar nebula are poor in deuterium. On the other hand meteoritic and interstellar organic molecules are enriched in deuterium. ADAPTED FROM ROBERT (2001)

values ranging from +5.5‰ to +22.9‰ and δD ranging from +1117‰ to +1388‰, whereas Murchison $\delta^{13}C$ values range from +19.1‰ to +28.1‰ and δD values from +389‰ to +1551‰ (Pizzarello and Huang 2002). As interstellar organic molecules are rich in deuterium, the D enrichment of those meteoritic organic molecules is consistent with interstellar precursors (FIG. 6). Alternatively, carboxylic acids may be formed in the meteorite parent body via the Strecker-cyanohydrin reaction (FIG. 4). When hydrogen cyanide and aldehydes are present, if the pH is acidic (pH <5) and if the abundance of ammonia is low, then α-hydroxycarboxylic acids are produced. On the other hand, if the pH is higher (close to neutral) and ammonia is present, then α-amino acids are formed. The ratio of α-hydroxycarboxylic acids to α-amino acids in a meteorite can thus provide information regarding the conditions on its parent body (volatile content and pH) (FIG. 4).

Nucleobases (Purines and Pyrimidines)

Several research groups have detected nucleobases in carbonaceous meteorites (Martins et al. 2008). Significant quantitative and qualitative variations, even among different fragments of the same meteorite, suggest that the observed nucleobases were produced by terrestrial contamination. As nucleobases are not chiral, stable isotope ratios of these compounds are necessary to establish their origin in meteorites. Compound-specific isotope measurements of nucleobases in carbonaceous meteorites were performed for the first time by Martins et al. (2008), using an extraction and purification procedure on a sample of the Murchison meteorite, followed by compound-specific carbon isotope measurements using gas chromatography–combustion–isotope ratio mass spectrometry. Martins et al. (2008) obtained $\delta^{13}C$ values for meteoritic uracil and xanthine of +44.5‰ and +37.7‰, respectively. These values unambiguously confirm a non-terrestrial origin for these compounds, and they demonstrate that components of the genetic code in modern biochemistry were produced abiotically in the early Solar System and may have played a key role in life's origin.

While meteoritic nucleobases are thought to be formed by abiotic synthetic pathways in a variety of cosmic environments, this class of compounds is vulnerable to UV photolysis in the interstellar medium and circumstellar environments (Peeters et al. 2003). Their synthesis is more probable in asteroid interiors, and several pathways have been proposed (Martins et al. 2008). Furthermore, the meteoritic distribution may be the result of synthesis and degradation producing further nucleobase species.

Polycyclic Aromatic Hydrocarbons

Although polycyclic aromatic hydrocarbons (PAHs) do not have a role in biological systems, Ehrenfreund et al. (2006) proposed that they may have served as scaffolds for protocells and played an important role in the structural development and organization of life. PAHs are the principal carrier of gas-phase carbonaceous material in space. Elsila et al. (2005) analysed the free organic components of 20 carbonaceous chondrites. They found that, for the CM2 meteorites analysed, the higher relative abundances of alkylated PAHs correlate with more intense aqueous activity in the meteorite parent body. In addition the thermally metamorphosed CK chondrites have no detectable levels of PAHs, suggesting that increased metamorphic intensity reduces the abundance of all PAHs. These molecules have also been identified in many other carbonaceous chondrites (Naraoka et al. 2000; Botta et al. 2008). A-881458 contains more than 70 PAHs, pyrene and fluoranthene being the most abundant. On the other hand, LAP 03784 contains several core PAHs (phenantherene/anthracene and pyrene). An extraterrestrial origin for the free aromatic components present in carbonaceous chondrites is indicated by large values for $\delta^{13}C$ (Sephton 2002).

SUMMARY AND CONCLUSIONS

Chyba and Sagan (1992) suggested that 3.8 to 4.5 billion years ago, during the period of heavy bombardment, extraterrestrial organic molecules were exogenously delivered to the early Earth. Therefore, carbonaceous meteorites, together with comets and interplanetary dust particles (IDPs), may have contributed the first prebiotic building blocks of life to our early planet. Laboratory analyses of meteoritic organic material, together with space missions to Solar System planets and satellites, asteroids[1] and comets[2], provide crucial insights into the early history of organic matter in our Solar System, the link between meteorites and their parent bodies, and the origin and evolution of life on our planet and elsewhere.

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1 The Hayabusa spacecraft returned to Earth in June 2010 after possibly collecting samples from the near-Earth asteroid 25143 Itokawa.

2 The Stardust spacecraft returned samples from comet Wild 2, and the Rosetta spacecraft is expected to reach comet 67P/Churyumov-Gerisimenko in 2014.

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