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Recognizing life in the Solar System: guidance from meteoritic organic matter

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Abstract: In the next decade, numerous space missions will attempt to detect organic matter on planets and other objects in the Solar System. Recognizing carbon-based life or its remains will be a fundamental goal of future missions. In preparation, studies of organic matter in meteorites are enabling scientists how to discriminate between biogenic and abiogenic materials.

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Introduction

Understanding the origin and uniqueness of life on Earth is fundamental to appreciating our place in the Universe. A key question is whether or not the Earth has been remarkably fortuitous in its amenable place in our Solar System and Galaxy, or if life on Earth is a logical consequence of the right mixture of energy, raw materials and chemical evolution. If the latter is true, then the chances that life may have begun elsewhere in the Universe are much higher. Searching for past or present life in our Solar System is one means of discriminating between the two possibilities.

Testing for life beyond the Earth is planned in the form of space missions to planets and moons in our Solar System with potential habitats for organisms. On Mars, NASA's Mars Exploration Rovers (Squyres *et al.* 2004) and ESA's Mars Express orbiter (e.g. Hauber *et al.* 2005; Murray *et al.* 2005) are currently setting the stage for future, more sophisticated vehicles that will carry instruments to search for molecular evidence of past life. In addition, NASA's Galileo space probe has revealed features on the icy surface of Jupiter's moon, Europa, that intimate the possibility of subsurface volcanic activity and the presence of liquid water below the ice crust (Carr *et al.* 1998). Future missions to Europa will investigate any subsurface ocean for alien biota and their organic remains.

Meteorites and organic matter

Whether these missions deliver unambiguous results will depend not only on their successful acquisition of data, but also on our ability to distinguish biogenic molecules from their cosmically widespread abiogenic counterparts (Ehrenfreund & Charnley 2000). Fortunately, to aid us in this objective, meteorites provide authentic extraterrestrial abiogenic material for scientific scrutiny. Meteorites are pieces of asteroids, which are Solar System objects many of which failed to grow large enough to ever experience 'planetary' processes such as core formation, volcanism or plate tectonics. The chondrite meteorites have near-solar volatile composition and radiometric ages of 4.566 billion years, reflecting their formation during, or shortly after, the birth of the Solar System (Allègre *et al.* 1995). Within the chondrites, there exists a carbonaceous subclass that contains significant amounts of organic carbon, a characteristic that contributes to a generally dark appearance. The chemistry and mineralogy of carbonaceous chondrites suggests that any organic compounds they contain should be ancient, predate the origin of life on Earth, and represent a perfect examples of molecules produced without biochemistry.

Much of our current understanding of meteoritic organic matter comes from investigations of a meteorite, which fell on 28th September 1969 near the town of Murchison, Australia (Fig. 1.). Other meteorites in the carbonaceous chondrite subclass are the famous Orgueil meteorite, which fell in France in 1864, and the Cold Bokkeveld meteorite that fell in South Africa in 1838. Yet the information derived from Murchison greatly surpasses that from the preceding carbonaceous chondrite falls because it arrived two months after the first lunar landing. Murchison was quickly delivered to several laboratories that had painstakingly developed methods to look for indigenous organic compounds in the Apollo lunar samples.

Interestingly, the recognition of indigenous organic matter in the fresh Murchison sample, alongside a growing awareness that the Earth's early atmosphere contained too small a proportion of reducing gases for the large-scale *in situ* production of organic matter (Kasting 1993), led to support

Table 1. *Types of mostly abiogenic organic matter in the Murchison (CM2) carbonaceous chondrite and their abundances*

Compounds	Abundances %	Abundances μg g^{-1} (ppm)	Reference
Macromolecular material	1.45		(Chang *et al.* 1978)
Carbon dioxide		106	(Yuen *et al.* 1984)
Carbon monoxide		0.06	(Yuen *et al.* 1984)
Methane		0.14	(Yuen *et al.* 1984)
Hydrocarbons:			
aliphatic		12–35	(Kvenvolden *et al.* 1970)
aromatic		15–28	(Pering & Ponnamperuma 1971)
Acids:			
monocarboxylic		332	(Lawless & Yuen 1979; Yuen *et al.* 1984)
dicarboxylic		25.7	(Lawless *et al.* 1974)
α-hydroxycarboxylic		14.6	(Peltzer *et al.* 1984)
Amino acids		60	(Cronin *et al.* 1988)
Alcohols		11	(Jungclaus *et al.* 1976b)
Aldehydes		11	(Jungclaus *et al.* 1976b)
Ketones		16	(Jungclaus *et al.* 1976b)
Sugar-related compounds (polyols)		~24	(Cooper *et al.* 2001)
Ammonia		19	(Pizzarello *et al.* 1994)
Amines		8	(Jungclaus *et al.* 1976a)
Urea		25	(Hayatsu *et al.* 1975)
Basic N-heterocycles (pyridines, quinolines)		0.05–0.5	(Stoks & Schwartz 1982)
Pyrimidines (uracil and thymine)		0.06	(Stoks & Schwartz 1979)
Purines		1.2	(Stoks & Schwartz 1981b)
Benzothiophenes		0.3	(Shimoyama & Katsumata 2001)
Sulphonic acids		67	(Cooper *et al.* 1997)
Phosphonic acids		1.5	(Cooper *et al.* 1992)

Fig. 1. The Murchison meteorite: a carbonaceous chondrite.

for proposals that the raw materials for life may have been delivered by extraterrestrial objects (Oró 1961). Further endorsement of an extraterrestrial source for the molecular precursors for biochemistry arose from the discovery that carbonaceous chondrites contain several classes of compounds that are important components in terrestrial organisms (Table 1).

Abiogenic organic signatures

Yet, within the meteoritic compound classes these potential forerunners of life exhibit chemical signatures that clearly identify them as being abiogenic in nature. The almost complete diversity in structure, a general decrease in abundance with increasing carbon number (i.e. molecule size), the presence of many compounds not used by terrestrial organisms and highly unusual stable isotope ratios are unambiguous signals that these molecules were synthesized under conditions that can only occur outside the Earth. For scientists interpreting future space flight data, searching for characteristics similar to those found in meteoritic molecules will be an essential first step in deciding whether or not life has been detected.

Amino acids

The first compound class usually associated with life, that was unambiguously identified and quantified in Murchison, was the amino acids (Kvenvolden *et al.* 1970) and to date more than 80 different amino acids have been found in this meteorite. Of these, only a few are the same as those utilized by terrestrial organisms. Eight α-amino acids (glycine, alanine, aspartic acid, glutamic acid, valine, leucine, isoleucine and proline) are constituents of proteins, while a few others such as β-alanine, α-aminoisobutyric acid (AIB) and sarcosine have a very restricted biological occurrence on Earth, for example as components of bacterial cell walls. The remainder are found naturally only in meteorites (i.e. they can exist on Earth, but have to be synthesized in the laboratory). Structurally diverse assemblages of amino acids are, it seems, a clearly abiogenic feature.

An important molecular signature of life is hosted by amino acids, namely the almost exclusive occurrence of only one (the left-handed) of the two possible enantiomers in all biosynthesized molecules. An enantiomer is one of the mirror image structures that exist for every molecule with an asymmetrically substituted (or chiral) carbon centre. The abiotic synthesis of chiral molecules always yields a 1:1 mixture of the right- (D) and left- (L) handed enantiomers, which is then called *racemic*. As meteoritic amino acids are synthetic products of abiotic processes, it is expected that they should be present as racemic mixtures. Indeed, racemic proportions were confirmed by early measurements of meteoritic amino acids such as alanine and isovaline (Kvenvolden *et al.* 1970, 1971). Recent re-examinations have verified the racemic nature of alanine but the same studies have uncovered slight differences in the abundance of the two enantiomers, termed enantiomeric excesses or '*ee*', of several amino acids in Murchison and the chemically-similar Murray meteorite (Cronin & Pizzarello 1997; Pizzarello & Cronin 2000). The L-enantiomer of the two forms of 2-amino-2, 3-dimethylpentanoic acid (DL-α-methylisoleucine and DL-α-methylalloisoleucine) as well as isovaline have an *ee* of about 8%. Both of these amino acids belong to the α-methyl amino acid family (i.e. methyl, amino and carboxyl groups are all attached to a common carbon atom) and have only a limited biological occurrence, making a terrestrial contamination source highly unlikely.

Immediately after recognizing *ee* in meteoritic molecules scientists deliberated the possible role of extraterrestrial amino acids in the formation of the first 'bio'polymers on the early Earth and the ultimate source of the chiral preference. In contrast to α-H amino acids (i.e. a hydrogen, amino group and carboxyl group are all attached to a common carbon atom) such as alanine, which, owing to the chemical lability of the α-hydrogen atom, could lose their *ee* in aqueous solution by racemization, α-methyl amino acids are enantiomerically stable and could therefore pass on this preference to other molecules during chemical evolution (Pizzarello & Weber 2004). The mechanism of such a chirality transfer is currently unknown, but several chemical model systems have dramatically demonstrated the potential of enantiomeric amplification during which a small excess is magnified by subsequent reactions (e.g. Soai *et al.* 1995, 2000; Sato *et al.* 2003). It is intriguing that the enantiomeric preference of the α-methyl amino acids in meteorites and the preference of the protein amino acids in terrestrial biology are the same and the role of small *ee* in precursor compounds for the origin of life on Earth or other planets is a fascinating area of investigation.

In our quest to discriminate life from non-life sources of organic matter in extraterrestrial environments, the interpretation of amino acid data from spacecraft missions must take into account the preference of life for certain amino acids and the apparent abiogenic *ee* in α-methyl amino acids. Consequently instruments designed for the search for amino acids on the surface or subsurface of Mars need to be able not only to detect and characterize individual amino acids at very low abundances, but they must also have the critical capability to separate and accurately quantify the enantiomers of chiral amino acids (e.g. Skelley *et al.* 2005).

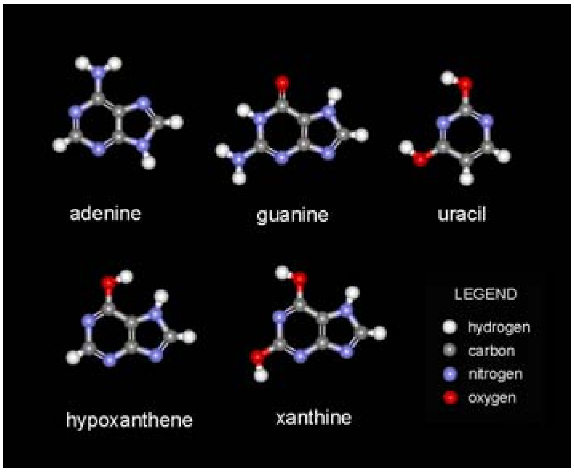

Fig. 2. Murchison nucleobases and related compounds. Similar units help host the genetic code in terrestrial life.

Nucleobases

The central role in biology for storage of genetic information is carried out by the nucleic acids, specifically the deoxyribonucleic acid (DNA) molecule. DNA and its sister molecule ribonucleic acid (RNA) are polymers comprised of monomeric units called nucleotides, which themselves are made out of three components: a nucleobase, a sugar and a phosphate residue. Compounds related to the first two classes are also found in carbonaceous chondrites. Nucleobases are one- or two-ring aromatic molecules that have one or several carbon atoms from the ring replaced by nitrogen atoms (Fig. 2.). Examples are adenine, guanine and uracil. These compounds, along with the structurally related compounds xanthine and hypoxanthine, were discovered in three carbonaceous chondrites (Murchison, Murray and Orgueil) in abundances about ten times lower than the amino acids (Stoks & Schwartz 1979, 1981a). Although the indigeinity of these molecules is still open to question their structural diversity implies an abiotic origin and provides some means of distinguishing them from their more structurally restricted biological counterparts. Once again assessing structural diversity will be important when interpreting experimental data from this biologically useful class of molecule.

Sugar-related compounds

Sugars are another component of nucleic acids, but this compound class has additional roles in biology. The storage of energy and the provision of structural support in organisms are roles undertaken by sugars of varying complexity. Sugars and their related compounds are collectively called polyhydroxylated compounds or polyols, reflecting their structure where a number of hydroxyl groups are attached to a carbon skeleton and carbonyl group. Abiogenic polyols are readily produced by the thermal polymerization of formaldehyde and this reaction, known as the formose reaction, results in a random distribution of a large number of polyols.

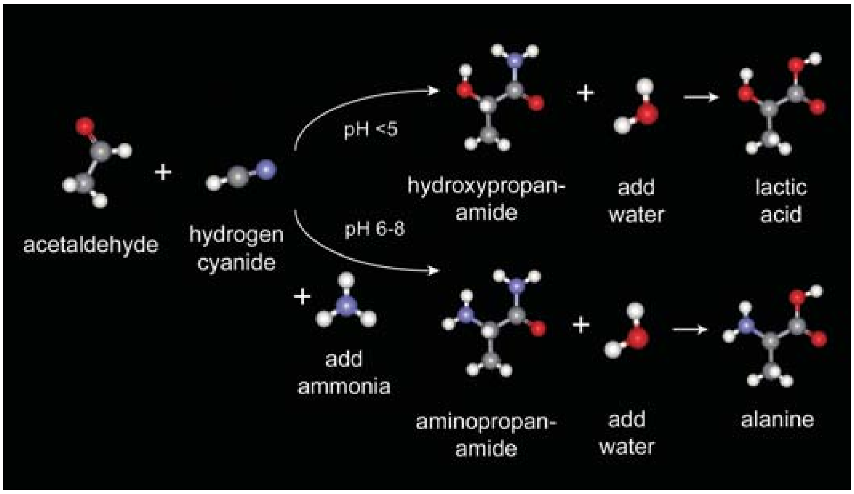

Fig. 3. The Strecker synthesis, a reaction that may have produced amino acids and hydroxyacids on asteroids in the early Solar System.

The fact that glycoaldehyde is abundant in the interstellar medium (Hollis *et al.* 2004) provides support for the notion that significant amounts of this sugar precursor compound were present in the planetesimals from which the planets and asteroids formed. Furthermore, several sugar-related compounds (sugar alcohols, sugar acids) and the simplest sugar molecule, dihydroxyacetone, were detected in Murchison (Cooper *et al.* 2001). For two compounds (glycerol and glyceric acid) abundances were determined on the same order of magnitude as for the amino acid glycine. Yet the simplicity and diversity of meteoritic polyols contrasts sharply with the high selectivity for their counterparts in terrestrial organisms, where deoxyribose and ribose are present in DNA and RNA respectively, glucose is the principle energy source and the highly ordered polysaccharide cellulose is the most abundant organic molecule in the biosphere. Consequently, the two sources are not likely to be confused. However, polyols pose significant analytical challenges and, because of the power and mass constraints of a planetary lander mission, current technology does not have the capability to extract and detect complex examples *in situ*. Therefore, a sample return mission would be the most appropriate approach for the determining an abiogenic or biologenic provenance of extraterrestrial sugar-related components.

Carboxylic acids

Compartmentalization is probably one of the most important properties of life, since it is one of the mechanisms to concentrate reactants from a dilute solution thereby allowing biochemical reactions to perpetuate. Amphiphilc molecules (those with a 'water-loving' and a 'water-hating' end) are key players as the principal components in all biogenic membranes, and long-chain carboxylic acids are one of the simpler classes of this type of compound. Yet, it has been shown that an organic acid fraction of a Murchison meteorite extract can form vesicles in an aqueous phase and although several components probably participated in the formation of the vesicle membranes, one of the components identified was nonanoic acid, a nine-carbon carboxylic acid (Deamer 1985, 1997). Investigations have shown, however, that the most abundant carboxylic acids in Murchison contain between two and five carbon atoms and display complete structural diversity, a general decrease in abundance with increasing carbon number, and an equal concentration of branched and straight-chain isomers (Lawless 1973; Yuen & Kvenvolden 1973). Overall, the generally small size and great structural diversity of meteoritic carboxylic acids is unlike the preference for very long carbon chains that are so useful in membrane formation in biological organisms and again the abiogenic and biogenic sources are relatively easy to distinguish.

Perhaps one firm indicator for abiogenic synthesis is forthcoming by direct comparisons of a particular, less abundant, class of carboxylic acid with the amino acids. The α-hydroxycarboxylic acids (or hydroxyacids) in carbonaceous chondrites contain up to nine carbon atoms and are structurally diverse (Peltzer & Bada 1978). Structural similarities between the hydroxyacids and amino acids suggest that the hydroxyacids and α-amino acids may be molecular siblings born on the parent asteroid and related by the *Strecker-cyanohydrin* synthesis. Hence similarities between these two compound classes are strong indictors of abiotic organic chemistry.

Polycyclic aromatic hydrocarbons

Undoubtedly the most dominant molecules in carbonaceous chondrites are the aromatic or polycyclic aromatic hydrocarbons (PAH) (Fig. 4.). When all organic size and solubility fractions are taken into account, PAH units constitute over 60 % of the organic matter in Murchison, the vast majority of which is present as an intractable macromolecular component. This molecular dominance should come as no surprise when it is recognized that PAH are the most abundant free organic molecules in space (d'Hendecourt & Ehrenfreund

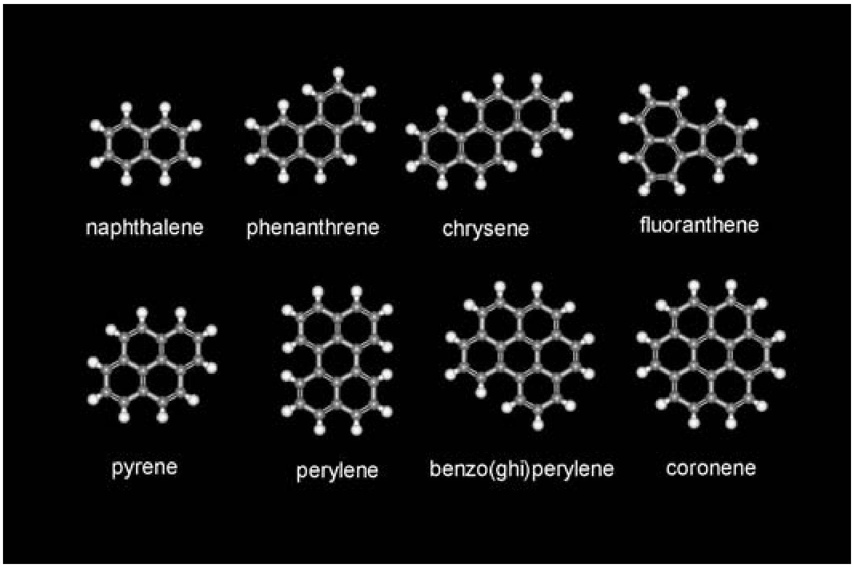

Fig. 4. Murchison PAH, the most abundant type of extraterrestrial organic compound in both meteorites and space.

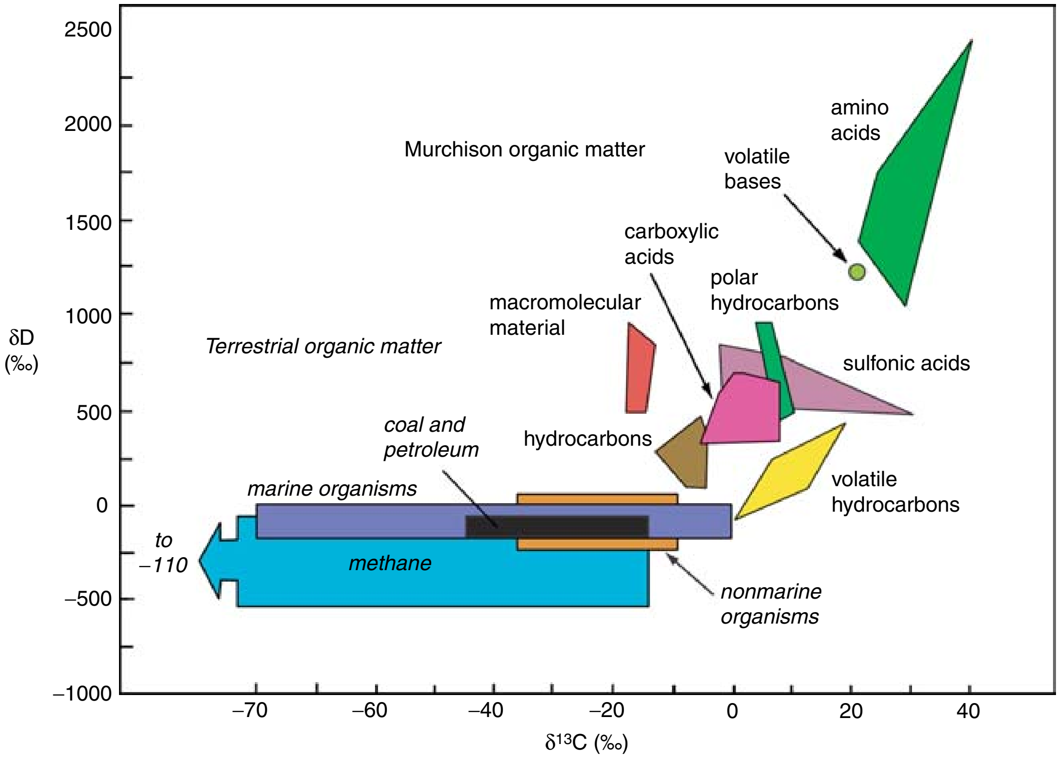

Fig. 5. The distinction between stable carbon and hydrogen isotope ratios in Murchison and life. The difference allows abiogenic extraterrestrial organic matter to be distinguished from its terrestrial biological counterpart. The abundances of stable isotopes are expressed using the δ notation. These indicate the difference, in per mil (‰), between the relevant ratio in the sample and the same ratio in an international standard as follows: $\delta‰ = ((R_{sample} - R_{standard})/R_{standard}) \times 1000$. Where $R = D\ (^2H)/^1H$ for hydrogen and $^{13}C/^{12}C$ for carbon. Data from Sephton (2002); Butterworth *et al.* (2004) and references therein.

1997). PAH are observed ubiquitously in our Galaxy as the carriers of the near- and mid-infrared (IR) bands (Tielens *et al.* 1999), and they are probably by far the most abundant molecular species in the diffuse interstellar medium. Moreover, multiple PAH emission features have been detected recently in other galaxies (e.g. Yan *et al.* 2005). PAH are thought be mainly produced in space in the high-temperature, high-density ejecta of asymptotic giant branch (AGB) stars (Cherchneff *et al.* 1992) and are the molecular intermediaries in the soot formation process.

Fig. 6. Image of the iron meteorite, dubbed 'Heat Shield Rock', discovered by the Mars Exploration Rover *Opportunity* on the surface of Meridiani Planum, Mars. This discovery illustrates that the accumulation of extraterrestrial material on the Martian surface could be significant (image courtesy of NASA/JPL/Cornell, see also http://marsrovers.jpl.nasa.gov).

In contrast to compound classes such as amino acids, nucleobases, polyols and carboxylic acids, PAH do not have a direct role in terrestrial biochemistry. PAH can be produced from biological organic matter only through partial combustion or thermal maturation following burial in the subsurface. PAH, therefore, are excellent indicators of abiogenic matter and can support interpretations that imply the absence of a biological contribution to organic mixtures. When found in close association with PAH, other less thermally stable compounds, such as those discussed above, should also be carefully examined for abiogenic signatures.

Perhaps the most famous example of extraterrestrial PAH being used to discriminate between biogenic and abiogenic processes is that of the Martian meteorite ALH 84001 in which three- to six-ring, predominantly parental (side-chain free), aromatic compounds were detected. When considering the relevance of the PAH alongside additional mineralogical phenomena, McKay *et al.* (1996) interpreted these molecules as the fossil remains of biological activity on Mars. The scientific community were unconvinced because parental PAH are not diagnostic of the many possible biotic and abiotic sources that could contribute organic matter for their formation (Anders *et al.* 1996). Ultimately, the PAH are better indicators of the environmental conditions in which abiotic processes operate to produce their final structure.

Stable isotope ratios

In addition to the very useful structural differences evident between abiogenic and biogenic molecules, the carbonaceous chondrite data reveal that extraterrestrial organic matter contains substantially elevated ratios for the stable isotopes of hydrogen (^{2}H(D)/^{1}H), carbon (^{13}C/^{12}C) and nitrogen (^{15}N/^{14}N) relative to life on Earth (Fig. 6.). Such data obtained from Murchison provided significant early evidence for an extraterrestrial abiogenic origin for meteoritic organic matter (Kvenvolden *et al.* 1970). Elevated D/H values clearly indicate a contribution from abiogenic interstellar molecules (Kerridge 1983; Yang & Epstein 1983) and methyl sulphonic acid in Murchison exhibits a relative enrichment in ^{33}S that can only be explained if the precursor of sulphonic acids was irradiated in the gas phase, most probably in interstellar space (Cooper *et al.* 1997).

The stable isotope distribution within molecular homologues is also evidently non-biological (Sephton 2002 and references therein). Short chain aliphatic hydrocarbons, amino acids, carboxylic acids and sulphonic acids all display ^{13}C/^{12}C ratio values that decrease with increasing carbon number, indicating a stepwise synthesis of higher molecular weight compounds from lower homologues. Moreover, meteoritic PAH exhibit an increase and then decrease in ^{13}C/^{12}C with increasing carbon number that is unlike anything observed in the combustion or maturation products of terrestrial organic matter.

The flux of extraterrestrial organic matter

With clear distinctions established between abiogenic and biogenic organic matter, it is useful to consider the likelihood of the former becoming a major component of the organic inventory of a planet and potentially confusing a mission designed to detect extraterrestrial biota. There are three main sources that can deliver organic molecules intact to planetary surfaces: asteroids (or their meteoritic fragments), comets and interplanetary dust particles (IDPs) (Chyba & Sagan 1992). Using the very recent history of the Earth as an example, the major mass, by about two orders of magnitude, is estimated to come from the IDP flux. The total flux of 'giant' micrometeorites (particles in the size range of 100 μm to about 1 mm) before atmospheric entry was determined directly from direct impact crater counts on the metallic plates of the Long Duration Exposure Facility (LDEF) satellite, from which a global value of $\sim(4\pm2)\times10^7$ kg yr^{-1} of extraterrestrial matter was inferred for this source (Love & Brownlee 1993; Maurette *et al.* 2000). Assuming a carbon content of $\sim$2.5 wt% (similar to carbonaceous chondrites), the annual accretion rate of carbon from micrometeorites and meteorites was estimated to be $\sim2.0\times10^5$ kg yr^{-1}, with meteorite-sized objects contributing a negligible 10^{-5} of that mass. Carbon delivery by comets is also relatively low and is thought to be three orders of magnitude less than for the IDPs both in modern and ancient times (Chyba & Sagan 1992).

Assuming minimal loss of carbon through atmospheric heating for these relatively small objects (Anders 1989) and taking a current biospheric carbon mass of 6×10^{14} kg, extraterrestrial delivery would rival Earth's biosphere in

3 Ga. Yet uncertainties exist, the amount of extraterrestrial infall was undoubtedly greater in the early Solar System and, although it is very difficult to scale the current flux of IDPs of today back to the early Earth, this flux may have been as high as 5×10^7 kg yr^{-1} at 4.0 Ga ago reducing the time required to 10 Ma. The number of meteorites, and presumably IDPs, accumulating on Mars is higher than on the Earth (×2.6; Shoemaker 1977; Bland and Smith 2000). Recent evidence of meteorite accumulation on Mars has been illustrated by the discovery of a meteorite within a 6 km traverse of the Mars Exploration Rovers (Fig. 6).

The proportion of meteoritic organic matter on planets such as Mars, therefore, is likely to be much higher than on the Earth, especially when the smaller relative abundance of indigenous biological organic matter (if any) on Mars is taken into account.

Studies of meteorites such as Murchison have empowered those scientists attempting to interpret whether extraterrestrial organic matter on other worlds is abiogenic or biogenic in origin. Despite sharing many compound classes, the organic matter in meteorites and biology are structurally and isotopically distinct. Yet, useful as these diagnostic features are, they also remind us of the great gap in our understanding of how prebiotic organic molecules can be transformed to the more complex, ordered and specific molecules utilized by terrestrial biology. Perhaps future data from less evolved organic systems on objects in our Solar System and beyond will reveal the path from chaotic chemistry to complex life.

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