Chapter

6

Water and Organics in Meteorites

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Abstract

The planet Earth incorporates tons of extraterrestrial materials on a daily basis. Meteorites represent some of the largest, most pristine (uncontaminated) and primitive (unaltered) specimens which offer glimpses of the early history of our solar system. Meteorites have brought to Earth not only water, but also a plethora of organic compounds, some of which are known to play an indispensable role in extant biology. Although these meteoritic molecules were chemically equivalent to those used in biology, they were synthesized in situ on the asteroidal parent bodies via complex chemical pathways, some of which require the presence of liquid water – another essential prerequisite for life. This chapter discusses the recent developments in our understanding of the water and organic inventories of meteorites, their possible origins, co-evolution, and final delivery to the early Earth.

Keywords:

Meteorite; Asteroid; Water; Organic matter; Prebiotic chemistry; Abiogenesis; Aqueous alteration

Introduction

Water and organic compounds are considered as the essential requirements for life. Water is a universal solvent that provides a medium for chemical reactions to take place. Carbon-based organic compounds made up of the element hydrogen (H), carbon (C), nitrogen (N), oxygen (O), phosphorus (P) and sulfur (S) form molecules with functions that are necessary for life.

While the high abundances of liquid water and organic compounds on the surface of Earth can sustain life, their presence is not limited to the Earth. Subglacial liquid water was detected below ice and dust layered deposits in Mars’ South pole using radar generated by the Mars Advanced Radar for Subsurface and Ionosphere Sounding (MARSIS) instrument onboard the Mars Express spacecraft (Lauro et al., 2021; Orosei et al., 2018). Enceladus, the sixth-largest moon of Saturn, has a global subsurface liquid saltwater ocean which ejects small organic precursors and complex macromolecular organic material via its cryovolcanic plume (cryovolcano is a type of volcano that erupts volatiles instead of molten rock) (Postberg et al., 2018; Thomas et al., 2016). Organic molecules which are vital for terrestrial biology, such as nucleobases and amino acids, have also been identified and confirmed to be indigenous to the hosting meteorites since the 1970s (Burton et al., 2012; Chan et al., 2018; Glavin et al., 2010; Kvenvolden et al., 1970; Martins et al., 2007) and cometary samples a couple of decades later (Altwegg et al., 2016; Elsila et al., 2009), however these biomolecules are not as ubiquitously distributed throughout the solar system as their smaller precursor molecules.

Water ice and organic molecules were accreted alongside other rocky components at the birth of our solar system to form planetesimals – building blocks of planets – from a spinning disc of dust and gas called the solar nebula. Despite the presence of water and organic precursors across the inner and outer regions of the solar system, early-accreted planetesimals from the inner solar nebula should have formed relatively dry as the nebular temperature in this region, where the early Earth located, was above the sublimation temperature of water ice (Lodders, 2003). Therefore, Earth’s water should have been delivered by later addition of volatile-rich planetesimals. These volatile-rich planetesimals were accreted beyond the “snow line” – which was at a distance of about 2−4 astronomical units (AU) in the early solar system (one AU is the average distance of the Earth from the Sun) – beyond which the nebular temperature was cold enough for water and organic compounds to condense as solids (Figure 6.1).

  Fig. 6.1 Water abundances of the atmosphere and bulk silicate (crust + mantle) of various solar system bodies. No Permission Required Water abundances of the atmosphere and bulk silicate (crust + mantle) of various solar system bodies. Data are from McCubbin and Barnes, 2019 and Rauf, et al, 2015

The main reservoirs of the volatiles are volatile-rich outer solar system materials such as comets, as well as carbonaceous asteroids that were formed in the outer region of the main asteroid belt. These outer solar system materials were perturbed by the inward and outward migration of giant planets, modelled by the “Grand Tack model” (Walsh et al., 2011), which swept and scattered outer solar system planetesimals into the inner solar system, enriching the formally volatile- and organic-depleted proto-Earth with water and organic material.

Life’s two key components are delivered to Earth every day through extraterrestrial materials since the birth of our solar system. Between 2,900 and 7,300 kg of extraterrestrial materials that are in the mass range of 10 g to 1 kg are delivered to the Earth’s surface every year in the form of meteorites (Bland et al., 1996). However, most of the extraterrestrial material are delivered in the form of tiny dust particles which are about 200 micrometres (µm) in diameter and 1.5 × 10-5 g in weight (Love & Brownlee, 1993; Rojas et al., 2021). These small particles are in the forms of micrometeorites (MMs) (~30–1,000 µm) (Genge et al., 2008) and interplanetary dust particles (IDPs) that are present as individual grains (5–15 µm) or as larger “cluster” particles (50–500 µm) (Brearley, 2006), and they contribute to approximately 4.7 × 106 kg of extraterrestrial materials per year.

The rate of extraterrestrial material delivery to Earth varied significantly over time. Evidenced by the impact crater record of our Moon, the impact rate was much higher at >3.8 billion years ago (Ga) than present, during a period known as the “heavy bombardment” (Bottke & Norman, 2017). The total flux of carbonaceous chondrites and biogenic elements delivered was at least five orders of magnitude higher during the “heavy bombardment” period compared to the present day (Pasek & Lauretta, 2008).

While meteorites are mainly fragments of asteroids, which are remnants of planetary formation, MM and IDPs are thought to have been derived from both asteroidal (Bradley et al., 1996; Carrillo-Sánchez et al., 2015) and cometary sources (Chan et al., 2020; Dobrică et al., 2012). Meteorites are further classified into three main categories: stony, iron, and stony-iron meteorites, based on their broad compositions of rock and metal. Within the stony category, a group of meteorites – the chondritic meteorites (constitute more than 80% of meteorites that reach the Earth surface) – has caught particular attention as they have not been melted since their formation, and thus are considered to retain crucial information about the early history of our solar system as it was formed.

About 4% of all recovered meteorite falls are carbonaceous chondrites that played a significant role in the origin of water and organic matter on Earth. Carbonaceous chondrites not only contain up to ∼3.5 weight total (wt.) % of carbon that is present in the form of organic compounds of diverse chemical and structural properties (Alexander et al., 2017), but also water (locked mostly as OH (hydroxyl) in clay minerals but also in the liquid form inside aqueous fluid inclusions), at the level of several percent, and in some cases up to 15% by mass (C. M. O’D. Alexander et al., 2017). Carbonaceous chondrites are further categorized into groups according to their chemical, isotopic and mineralogical characteristics to indicate that meteorites from the same group are derived from the same parent body (Weisberg et al., 2006). These carbonaceous chondrite groups are CI (Ivuna-like), CM (Mighei-like), CO (Ornans-like), CV (Vigarano-like), CK (Karoonda-like), CR (Renazzo-like), CB (Bencubbin-like), and CH (high iron; ALH 85085-like), with the second letter designated by the first initial of the typical meteorite specimen for that group.

In this chapter, we discuss the occurrences of water and organic matter in extraterrestrial materials, the different views of their formation origins, how were they delivered to the Earth’s surface, and how advanced analytical methods are used to distinguish indigenous (extraterrestrial) water and organics from terrestrial contamination, in order to extract the valuable information of their formation at the birth of our solar system.

Water in meteorites

Chondrites carry the best record of the history, distribution, and activity of water in the early solar system. Chondritic meteorites, especially carbonaceous chondrites, have special significance, being similar in composition to the solar photosphere excluding a few very volatile elements (Anders & Grevesse, 1989). These meteorites are direct samples of primitive asteroids and potentially comet nuclei materials. Water, as a solvent and mass transport agent, is probably the main source of low-temperature chemical alteration in chondrites. Oxidation, hydrolysis, and carbonation reactions resulting from aqueous fluids reacting with chondritic minerals are collectively termed aqueous alteration.

Hydrous mineral phases

The most thorough recent review of aqueous alteration in meteorites is given by (Brearley, 2006) and (Zolensky et al., 2018), which are references for much of what follows below. In most altered meteorites (and MMs and chondritic IDPs), the most direct evidence of aqueous alteration is the presence of phyllosilicates or pseudomorphs after same (Figure 6.2). The most abundant extraterrestrial phyllosilicates are saponite and serpentine family minerals (Figure 6.2a) including cronstedtite, chrysotile, and antigorite. Far less abundant are chlorites, micas and talc. CM chondrites usually contain the hydrous sulfide tochilinite (Figure 6.2b), intergrown with serpentine. Secondary assemblages associated with these phyllosilicates include diverse carbonates (mainly calcite, dolomite, magnesite and siderite), oxides (mainly magnetite and chromite), calcium phosphates (mainly apatite and whitlockite), Fe-Ni sulfides (pyrrhotite and pentlandite), and Fe-Ni carbides (cohenite ((Fe,Ni)3C) and haxonite ((Fe,Ni)23C6)). Fe-rich olivine (fayalite) may also form during aqueous alteration, and this fact is the basis for chronology of aqueous alteration (Doyle et al., 2015). The timing of aqueous alteration has also been measured using carbonates, but so far with inferior standards so those dates are tentative (its proven difficult to make the correct standards). 21

  Fig. 6.2 Back scattered electron (BSE) images of hydrous and secondary mineral phases in meteorites. No Permission Required Back scattered electron (BSE) images of hydrous and secondary mineral phases in meteorites. (a) Serpentine vein crosscuts fine-grained serpentine-saponite matrix in the Ivuna CI1 meteorite. (b) Grain of intergrown tochilinite (white) -serpentine (grey) in the Maribo CM2 meteorite. (c) Magnetite framboids in the Ivuna CI1 meteorite. (d) Ca carbonate is recognizable as calcite in the Sutter’s Mill meteorite because of the obvious twinning. Note black holes which are opened fluid inclusions.

The abundance of these secondary phases increases from Enstatite (E) chondrites (found in only the meteorite Kaidun, which is ~50% E chondrite), through ordinary chondrites: LL, L, and H, and then through the carbonaceous chondrites in this order: CO, CV/CK, CB/CH, CR, CM, CI. Carbonaceous chondrites as a class are characterized by nearly solar elemental abundances, high carbon content and volatile element enrichments, although these volatiles vary greatly between the different carbonaceous chondritic types. As its name indicates, the carbonaceous chondrites contain the highest carbon and water concentrations among chondritic meteorites (up to 3.7 wt.% of carbon and at least 5 wt.% of water in CI). Nevertheless, carbonaceous chondrites show a wide range in their textural characteristics, degree of oxidation, aqueous alteration, shock- and thermal metamorphism, due to complex and varying nebular and asteroidal histories.

In all but the CO and CM chondrites, saponite is the most abundant phyllosilicate. Interestingly, saponite is practically always present in association with serpentine, but saponite is lacking in CO and CM chondrites, possibly due to differences in alteration temperature or solution pH.

Liquid water inclusions

Despite widespread aqueous activity in the early solar system, actual preserved water samples in meteorites are very rare. A report of aqueous fluid inclusions in phosphates in the ordinary chondrite Jilin still requires confirmation (Ed Roedder, personal communication, 1999). Two ordinary chondrite regolith breccias, Monahans 1998 (H5) and Zag (H3-6) contain fluid inclusion-bearing halite (NaCl) crystals (Rubin et al., 2002; Zolensky et al., 1999), which originated on a carbonaceous body (possibly a P- or D-class asteroid) (Figure 6.3). The halites were dated by K-Ar, Rb-Sr and I-Xe systematics to be ~4.5 Ga and thus the trapped aqueous fluids are at least as ancient.

  Fig. 6.3 Water-bearing halite crystal in the Zag meteorite No Permission Required Water-bearing halite crystal in the Zag meteorite. (a) A blue halite crystal in the Zag meteorite. The halite crystal is about 2 mm across. (b) A cluster of fluid inclusions in a blue halite crystal of the Monahans meteorite. White arrows indicate vapour bubbles that moved freely in two fluid inclusions. These two fluid inclusions are about 5−10 µm in size.

Heating/freezing studies of these aqueous fluid inclusions demonstrated that they were trapped at low temperature, and recent investigation shows that the preserved brines contain abundant inorganic and especially organic species (Bodnar et al., 2019). (Tsuchiyama et al., 2021) reported CO2-bearing aqueous fluid (CO2/H2O > ~0.15) in a fluid inclusion in a calcite crystal in the Sutter’s Mill CM chondrite. These and the halites are direct evidence of dynamic evolution of the solar system, requiring the water-bearing parent asteroids to have formed outside the solar system’s snow line and later transportation to the inner solar system due, probably, to Jupiter’s orbital instability. Thus, compositional data on fluid inclusions in extraterrestrial phases have the potential to reveal unique information regarding the origin and activity of aqueous fluids in the early solar system, and especially their interactions with organic material.

Aqueous alteration on asteroid parent bodies

There is still controversy as to where aqueous alteration took place. It has been suggested that aqueous alteration of early condensates occurred as the solar nebula cooled in reactions between solid grains and water vapor, or, alternatively, after accretion in an asteroidal environment (Brearley, 2006). The effects of these parent body alteration processes are difficult to separate from alteration that may have taken place prior to accretion, but the preponderance of petrographic evidence suggests that the bulk of the alteration (possibly all) occurred after accretion of asteroids within those solid bodies as reactions with aqueous fluids and vapor. This view is supported by the presence of veins of secondary minerals crosscutting other lithologies (see Figure 6.2a), alteration haloes, consistent mineral compositional zoning across large volumes of sample, and similar observations.

The conditions for aqueous alteration in chondritic parent bodies varied significantly in terms of temperature (including temperatures below 0°C), pressure, water to rock ratio, redox conditions, fluid mobilization, and escape of volatiles (M. E. Zolensky et al., 2018). The study of aqueous alteration permits placing constraints on the volatile budget inherited from the solar nebula and the extent of chemical redistribution, changes in petrologic relationships, and isotopic fractionation that took place in chondritic parent bodies. This understanding of aqueous alteration has come very gradually (Brearley, 2006), as many meteorites with no obvious record of aqueous alteration have ultimately come to be recognized as having been thoroughly altered, then thermally metamorphosed to largely erase the record (Tonui et al., 2014). In many meteorites the record of aqueous alteration is preserved only as rare fragments of lithologies, preserved in regolith breccias of otherwise anhydrous materials (M. E. Zolensky, 2021).

Chondritic parent bodies have been heated to a variety of temperatures in different timescales. The consequences of thermal metamorphism are textural, compositional, and mineralogical homogenization, recrystallization, and loss of volatiles. As one example of post-aqueous alteration thermal metamorphism, of the known CI chondrites (the most volatile rich meteorites) half have been heated either by thermal metamorphism, solar heating, or impact heating, and in these stones phyllosilicates are replaced first by highly disordered and defect-rich structures and finally by olivine, and carbonates are replaced by oxides ((Tonui et al., 2014, 2014); Tomoki Nakamura, personal communication, 2016). Thermal metamorphism has been attributed (1) to electromagnetic induction by solar winds, (2) to decay of short-lived radionuclides such as 26Al in the parent body, (3) to heating during accretion or during later impact events (M. Zolensky & McSween, 1988), and (4) by orbits that carry asteroids near the Sun (Nakamura, 2005). In first three cases the metamorphism must have been completed very early in solar system history, but the fourth process could have happened at any time - even very recently. A number of evidences indicate that most thermal metamorphic features were caused by isotopic decay and impact-driven thermal/hydrothermal metamorphism. However, these metamorphic indicators often conflict with each other (Tonui et al., 2014). Our views of the causes of metamorphism will undoubtedly continue to evolve as new meteorites are studied, notably the samples just collected from asteroids Ryugu and Bennu (see Section "Sample return space missions").

Organic matter in meteorites

Organic analyses of meteorites have spanned a long history, however, the initial observations prior to the 1970s were frequently subject to doubt because of the possibility of terrestrial contamination. The bulk of our current understanding of the organic content of meteorites comes from the study of two large-mass carbonaceous chondrites, the Murchison and Allende meteorites, which fell to Earth in 1969. This was the same year when we celebrated the excitement of the return of the first lunar samples by the Apollo 11 mission. Early studies have detected small amounts of organic compounds in the Apollo 11 samples, such as hydrocarbons and amino acids (Harada et al., 1971; Hare et al., 1970; Nagy et al., 1970), however, these studies could not vigorously resolve the origin of the analytes as the early techniques could not resolve the chirality of amino acids nor provide compound-specific isotopic analysis. Nevertheless, the techniques and laboratories developed for the organic analyses of the lunar samples provided a timely capability to detect organic compounds present in meteorites (Kvenvolden et al., 1970).

Organic phases

Carbonaceous chondrites are among the most water rich materials in the inner solar system, containing up to ∼10 wt.% of water (C. M. O’D. Alexander et al., 2012), which is even higher than that estimated for the bulk silicate Earth (BSE – the silicate part of the Earth excluding the core; contains about 700–3,000 parts per million (ppm) of water) (Peslier et al., 2017). Carbonaceous chondrites can have high bulk carbon content that is composed predominantly of organic matter (3.5 wt.%), but also mineral phases such as carbonates (0.2 wt.%), diamond (0.04 wt.%), silicon carbide (0.009 wt.%), and graphite (0.005 wt.%) (C. M. O’D. Alexander et al., 2017; Amari et al., 1990; Grady et al., 1988; Lewis et al., 1987; Sephton et al., 2003; Smith & Kaplan, 1970) (Figure 6.4), some of which are presolar grains inherited from previous solar systems.

  Fig. 6.4 Carbon phases in carbonaceous chondrites. No Permission Required Carbon phases in carbonaceous chondrites. Data are from Glavin et al., 2018 and references therein.

Organic matter is generally classified into three main fractions: solvent-insoluble organic matter (IOM), solvent-soluble organic matter (SOM) and volatile components. SOM can be extracted from the meteorite by organic solvents of different polarities such as water, formic acid, methanol, dichloromethane, hexane, or their mixtures (Glavin et al., 2006) (Table 6.1). IOM can be isolated by HCl/HF or CsF/HF demineralization via the removal of carbonates and silicate minerals (Cody et al., 2002). Volatile compounds can be released by thermal extraction or supercritical fluid extraction (Belsky & Kaplan, 1970; Sephton et al., 1998).

  Table 6.1 Types of organic matter in extraterrestrial samples, their extraction methods, and C and N isotopic compositions.

Types of organic matter in extraterrestrial samples, their extraction methods, and C and N isotopic compositions. The table was compiled from the following references: Sephton, et al, 2003; Cronin, John R. and Chang, 1993; Sephton, et al, 1998; Sephton, M. A., et al, 2004

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
|  Types of organic matter  |  Extraction methods  |  % of total organic content  |  δ13C (‰)  |  δ15N (‰)  |  Examples of released molecules  |
| Soluble (SOM) | Free (FOM) | Organic solvents | 20-30 | +7 | +88 | Amino acids; amines; nucleobases; alcohols; polyols; carboxylic acids |
| Insoluble (IOM) | Labile (LOM) | Hydrous pyrolysis; Thermal degradation; Residue of HCl/HF or CsF/HF demineralization | >20 | ≤−5 | ≤+85 | Low molecular weight hydrocarbons, aldehydes, ketones, amines, alcohols, monocarboxylic acids |
| Refractory (ROM) | Residue of HCl/HF or CsF/HF demineralization | >50 | ≤−20 | ≤−25 | Complex network of macromolecular carbon of at least five- or six-ring polyaromatic hydrocarbon cross-linked together |

The majority of organic matter (>50% of total organic carbon) in meteorites is in the form of structurally complex IOM macromolecules (C. M. O’D. Alexander et al., 2017, 2007; Bonal et al., 2007; Busemann et al., 2007; Chan et al., 2018; Cody & Alexander, 2005; Kebukawa et al., 2019; Quirico et al., 2014; Yabuta et al., 2007) (Figure 6.4; Table 6.1).

IOM is considered “non-labile”, or “refractory” (Table 1), and is a kerogen-like polymer made up of a complex network of condensed small (smaller than C24) polycyclic aromatic hydrocarbons (PAHs), and heteroaromatic macromolecules that contain alkyl branching, functional groups such as OH and COOH, and are bridged by short, highly branched alkyl chains, ether, and sulfide linkages (Pizzarello et al., 2006; Yabuta et al., 2007). Although PAHs have no direct role in terrestrial biochemistry, they are the predominant component of IOM, and are recognised as the most abundant free organic molecules in space (D’Hendecourt & Ehrenfreund, 1997). The elemental compositions of IOM have been estimated to span a range of H/C ratios (which reflects the IOM’s structure such as aromaticity - higher aromaticity for lower H/C ratio) from C100H48N1.8O12S2 (H/C = 0.5) (Zinner, 1988) to C100H70N3O12S2 (H/C = 0.7) (Hayatsu et al., 1977).

SOM is made up of smaller macromolecules and simpler species such as amino acids, amines, nucleobases, alcohols, polyols, and carboxylic acids (Aponte et al., 2015; Burton et al., 2012; Chan et al., 2018; Cooper et al., 2001; Glavin et al., 2018; Martins et al., 2008; Pizzarello & Holmes, 2009) (Figure 6.5). These species may diffuse rapidly through pores in asteroidal materials (which can have porosities as high as 25% (Britt & Consolmagno, 2003)), and some may undergo chemical reactions with aqueous fluids at temperatures as low as 275K.

  Fig. 6.5 Soluble organic compounds and their organic structures. No Permission Required Soluble organic compounds and their organic structures.

Volatile compounds are light molecules such as CO2, light hydrocarbons (e.g. benzene, C1-C4 alkylbenzenes, C1 alkyl naphthalenes) and low molecular weight aldehydes, ketones, amines, alcohols and monocarboxylic acids (Basile et al., 1984; Belsky & Kaplan, 1970; Cronin & Chang, 1993; Krishnamurthy et al., 1992; Sephton, 2002; Sephton et al., 2004; Yabuta et al., 2007). They are easily lost during conventional solvent based preparation techniques, as the necessary solvent evaporation stages, such as drying under a stream of nitrogen gas, can remove compounds that are more volatile than the tricyclic polyaromatic hydrocarbons (Krishnamurthy et al., 1992). Therefore, only compounds which are “immobilized”, such as molecules that are locked within crystals, within macromolecular carbon, and adsorbed onto grain surfaces, can be preserved for analyses (Belsky & Kaplan, 1970; Tsuchiyama et al., 2021).

Extraterrestrial organics and their significance for terrestrial biology

Life is defined as any system that is “capable of reproduction, mutation and reproduction of its mutations” (Sagan, 1994). Carbohydrates, lipids, nucleic acids, and proteins are four major macromolecules that are essential to terrestrial life as we know it. These polymers are required in biology to carry out replication and catalysis. Therefore, without these components, life is impossible. Although these complex molecules have not been identified in extraterrestrial materials, their building blocks have been observed, presenting a wide range of structural complexities.

Extraterrestrial organic compounds can be synthesized abiotically through different mechanisms (Figure 6.6), such as (1) irradiation of interstellar ice (H2O, CO, CO2, CH4, CH3OH, NH3) in the interstellar medium (ISM) and circumstellar medium (CSM) via ultraviolet (UV) light (Bernstein et al., 2002; Munñoz Caro et al., 2002; Nuevo et al., 2012), (2) aqueous reactions involving liquid water on asteroid parent bodies (Cody et al., 2011; Kebukawa et al., 2019; Pizzarello & Weber, 2004), and (3) mineral surface-catalyzed reactions of precursor molecules (e.g. H2, CO, and NH3) at elevated temperatures via Fischer-Tropsch Type (FTT) synthesis (Hayatsu et al., 1971; Hill & Nuth, 2003; Pizzarello, 2012).

  Fig. 6.6 Examples of different abiotic organic synthetic pathways. No Permission Required Examples of different abiotic organic synthetic pathways.

Amino acids

Proteins are effective at catalyzing reactions with superior speed and specificity, with protein-catalyzed reactions being 10,000 to 100,000 times faster and more specific than ribonucleic acid (RNA) catalysis (Jeffares et al., 1998; Penny, 2005). Amino acids are the building blocks of proteins and enzymes that are essential for biology. Amino acids of C2−C10 in length have been identified in carbonaceous chondrites, from below a few parts per billion (ppb) to up to hundreds of ppm abundance, and they exhibit nearly complete structural diversity (all isomeric forms present) (Glavin et al., 2010; Koga & Naraoka, 2017; Martins et al., 2007; Pizzarello & Holmes, 2009) (Table 6.2).

  Table 6.2 Extraterrestrial organic compounds detected in meteorites and their total abundances, detected chain lengths, and carbon isotopic compositions.

Extraterrestrial organic compounds detected in meteorites and their total abundances, detected chain lengths, and carbon isotopic compositions.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  Organic compounds  |  Total abundance  |  Chain length  |  δ13C  |  References  |
| Amino acids | ~2500 nmol/g | C2−C10  | −25 to +26‰ |  |
| Nucleobases | ~4.5 nmol/g | -- | +38 to +45‰ |  |
| Polyols | 85 nmol/g | C2−C6  | +5 to +60‰ |  |
| Carboxylic acids | ~5934 nmol/g | C1−C12  | −57 to +44‰ |  |

Nucleobases

Nucleobases (substituted one-ring (pyrimidine) or two-ring (purine) nitrogen heterocyclic compounds, such as adenine, guanine, uracil), are found in the nucleic acids ribonucleic acid (RNA) and deoxyribonucleic acid (DNA) in terrestrial life, which play a central role in biology for storage of genetic information. RNA is also responsible for the information transfer of the genetic code from DNA to ribosome, which serves to link amino acids in correct sequence to form proteins.

Although nucleobases have been detected in carbonaceous chondrites since the 1960s (Hayatsu, 1964; Krishnamurthy et al., 1992; Martins et al., 2008; Shimoyam et al., 1990; Stoks & Schwartz, 1981; van der Velden & Schwartz, 1977), these early findings were subject to questions as it was difficult to completely rule out a possible origin as terrestrial contamination, as the observations were possibly interfered by co-eluting compounds that masked the isotopic signatures. However, with the use of ultra-high-resolution mass spectrometry, (Callahan et al., 2011) showed that about 1−244 ppb of each nucleobase was present in carbonaceous chondrites with a diversity distribution that is distinctive from terrestrial signatures.

Polyols

Another essential component of nucleic acids that has also been identified in meteorites is polyols (e.g. sugars, sugar alcohols, sugar acids). Sugars polymerize into large carbohydrates called polysaccharides. They play an important role in terrestrial life due to their capability of energy storage and the provision of structural support in organisms. A variety of C2−C6 sugar alcohols and sugar acids have been identified in carbonaceous chondrites at an individual abundance of 4−8,000 ppb (Cooper et al., 2001; Cooper & Rios, 2016), however, an extraterrestrial signature has been established for just a few of them (C3−C4 sugar acids) as limited by the sample mass.

Carboxylic acids

Carboxylic acids form fatty acids and lipids that are key components of the bi-layer cell membrane structure in biology. Aliphatic mono- and di-carboxylic acids up to C12 in length have been identified in carbonaceous chondrites, at total abundances of 10 to 100 times the amount of amino acids (Aponte et al., 2011; Huang et al., 2005; Krishnamurthy et al., 1992; Lawless & Yuen, 1979; Martins et al., 2006; Pizzarello & Huang, 2002; Yuen et al., 1984). The presence of extraterrestrial carboxylic acids has been confirmed by elevated heavy isotope enrichments in hydrogen and carbon isotopic analyses (Aponte et al., 2011; Huang et al., 2007).

The roles of water

Water plays an essential and yet complex role in the formation, modification, and destruction of organic material on asteroids. When the chondrite parent bodies warmed up from heat contributed by the decay of radionuclides, episode(s) of aqueous alteration took place in which the accreted interstellar organics underwent various reactions. Water can modify the morphology, isotopic signature, molecular structure, and spatial distribution of the organic matter.

The extent of thermal and aqueous alteration is reflected by the “petrologic type” of a meteorite (Huss et al., 2006; Weisberg et al., 2006). A number from 3 to 6 indicates increasing thermal metamorphism, and from 3 to 1 an increasing degree of aqueous alteration. This number is indicated after the meteoritic group. Therefore, a heavily aqueously altered CI type meteorite like Orgueil is referred as CI1. An increasing degree of aqueous alteration can result in increases in aromaticity of IOM and a depletion in heavy stable isotopes of carbon and nitrogen, as the organics enriched in heavy isotopes can be liberated during aqueous alteration (C. M. O’D. Alexander et al., 2014; Sephton et al., 2003).

Organic compounds can be synthesized by abiotic processes (Figure 6.6 and Figure 6.7). The SOM fraction, including amino- and hydroxy-acids, for example, can be synthesized by Strecker-cyanohydrin type synthesis, Michael addition of ammonia to α,β-unsaturated nitriles, and HCN polymerization through aqueous processing (Ehrenfreund et al., 2001; Kebukawa et al., 2017; Lerner et al., 1993; Peltzer et al., 1984). Straight-chained monocarboxylic acids can be produced by hydrolysis and reduction of alkyl nitriles (e.g., methyl and ethyl cyanide).

  Fig. 6.7 A simplified schematic of synthetic relationships between extraterrestrial SOM From Simkus, D. N., Aponte, J. C., Elsila, J. E., Parker, E. T., Glavin, D. P., & Dworkin, J. P. (2019). Methodologies for analyzing soluble organic compounds in extraterrestrial samples: Amino Acids, Amines, Monocarboxylic Acids, Aldehydes, and Ketones. Life, 9(2). https://doi.org/10.3390/life9020047 A simplified schematic of synthetic relationships between extraterrestrial SOM including (1) amines, (2) amino acids, (3) aldehydes/ketones, and (4) monocarboxylic acids. The listed synthetic pathways are: amine synthesis via (i) reductive amination of aldehydes/ketones, (ii) decarboxylation/deamination of amino acids; Amino acid synthesis via (iii) Strecker reactions and (iv) CO2 addition to amines; and Carboxylic acid synthesis via (v) oxidation of aldehydes. Other potential mechanisms for these molecules are not shown here. Modified from Simkus, et al, 2019.

However, prolonged aqueous alteration may result in the decomposition of the synthesized organics through interactions with liquid water and minerals. Extended aqueous and thermal processes can decompose amino acids through deamination and decarboxylation to form carboxylic acids and amines (Aponte et al., 2011; Cohen & Coker, 2000; Pietrucci et al., 2018; Rodante, 1992) (Figure 6.7). This accounts for the lower amino acid abundances observed for the more aqueously-altered meteorites compared to the less aqueously altered ones (Glavin et al., 2010; Glavin & Dworkin, 2009).

Temperature is shown to be a critical factor in determining whether organics are synthesized or decomposed during aqueous alteration. Aqueous alteration at low temperatures (<120 °C), like that experienced by CI and CM chondrites (R. Hayatsu et al., 1977), results in the preferential synthesis of α-amino acids and straight-chain monocarboxylic acids. In contrast, in carbonaceous chondrites that have been exposed to extensive aqueous alteration as well as short-term heating at elevated temperatures up to 400−600ºC (e.g. CI-like Yamato (Y) 980115, Wisconsin Range (WIS) 91600, sometimes now called “CY” chondrites), there is a depletion of straight-chain relative to branched monocarboxylic acids, and a domination by straight-chain n-ω-amino acids (Aponte et al., 2011; Burton et al., 2012; Chan et al., 2016).

Water also imposes indirect influence onto the meteoritic organic content by promoting the formation of secondary minerals (e.g. clays, carbonates, salts) that can catalyze the formation of organic compounds, and thus can trap, concentrate, and protect adsorbed organic compounds, thereby promoting polymerization reactions (Chan et al., 2018, 2017; Le Guillou et al., 2014; Pearson et al., 2002; Pizzarello, 2012). Another important role of water is to provide a mechanism for chiral symmetry breaking that kick started the chiral “preference” of amino acids and sugars observed in terrestrial biology. This will be discussed in further detail in Section "Enantiomeric ratios".

Delivery of meteorites

Space weathering

Despite thousands of spectral measurements of meteorites under quite varied conditions, and thousands of reflectance spectra of asteroids, there are very few satisfying matches between meteorites and asteroids (Greenwood et al., 2020), despite the fact that we know the former must derive from the latter. For example, spectra of S-type asteroids (the most common type of near-Earth asteroid) exhibit weakening of absorption bands and reddening relative to spectra of ordinary chondrites which derive from them. These spectral mismatches are explained by “space weathering”, where the primary known mechanism of such spectral change is production of nanophase metallic iron particles, as shown by studies of the amorphous rims of lunar soil grains (Keller & McKay, 1993). Vapor-deposition during impacts by high-velocity IDPs, and implantation of solar wind ions are responsible for the space weathering rims. Indeed, regolith grains from S-class asteroid Itokawa gathered by the Hayabusa spacecraft revealed nanometer-thick, amorphous space-weathered rims containing nanophase iron and sulfur (Noguchi et al., 2011). Some of the space-weathered rims contain vesicles formed by trapped solar wind helium. Such thin amorphous rims can be developed in timescales of 1,000 years (based on solar flare density and implanted noble gas concentrations).

Can similar effects be expected from regolith on hydrous asteroids? Although there is as yet no ground truth regarding space weathering of hydrous materials, this situation is about to change as we get our first look at the Ryugu samples in the latter half of 2021. C-class asteroids like Ryugu frequently exhibit reflectance spectra consistent with thermally metamorphosed carbonaceous chondrites (Vilas, 2008). It is usually held that hydrous asteroids would generally be disrupted by rather than preserving evidence of impact processing. Nevertheless, even the water-rich CI and CM chondrites exhibit evidence of impact shock (M. Zolensky et al., 2015), especially brecciated carbonaceous chondrites.

We identified agglutinate grains (soil grains aggregated in a vesicular glassy matrix) in the carbonaceous chondrites Orgueil (CI1), Nogoya and Diepenveen (both CM2) (Figure 6.8). These agglutinates contain devitrified, melted matrix grading into desiccated phyllosilicates. The cooling melts partially devitrified into normally-zoned olivine crystals which sit in mesostasis glass. It is interesting that these glasses have partially devitrified to olivine, in contrast to the situation for lunar agglutinates (Lindsay Keller, personal communication, 2014). This could be due to a higher olivine normative composition, and the difficulty of quenching a liquid with an almost pure olivine composition (Gary Lofgren personal communication, 1999). The melted portion of the chondrite agglutinate grains grades gradually into very porous, dehydrated phyllosilicates. The pores were undoubtedly formed by volatile (water and sulfur) loss during heating and dehydration. Troilite and Fe-Ni metal fill fractures in olivine crystals. The fact that the sulfides were not completely volatilized in these samples requires rapid heating and quenching.

  Fig. 6.8 BSE images of a fragmented agglutinate in the Nogoya CM2 meteorite. No Permission Required BSE images of a fragmented agglutinate in the Nogoya CM2 meteorite. (a) Image of the entire agglutinate fragment. Close-up images of (b) and (c) are marked by the upper and lower yellow boxes, respectively. (b) Close-up of the agglutinate grain, where complete melting has occurred, and cooling has produced normally-zoned olivine crystals in mesostasis glass. (c) Close-up of partially dehydrated matrix at the edge of the agglutinate. Arrows indicate dehydration pores and cracks among the matrix phyllosilicates.

In the same carbonaceous chondrites there are sub mm-sized beads of glass which are most likely additional products of impact, as well as masses of melted sulfides recording flash heating events (impacts). The troilite masses contain matrix silicate grains which must have been injected into them while the sulfides were still molten. Melted sulfides are also found in shock veins in carbonaceous chondrite lithologies in the Kaidun meteorite (M. Zolensky & Ivanov, 2003).

Since meteorites cannot fully represent asteroid regolith, we propose that such shock-impact-altered material is far more abundant in poorly-indurated regolith. Thus, these materials will likely be present in significant quantity on the surfaces of C-class asteroids being sampled by the Hayabusa2 and OSIRIS-REx spacecrafts.

Grand Tack

The H2O snow line was not a sharp boundary (Figure 6.9). Properties of the dusty gas cloud varied with time due to changes in the brightness of the Sun, the density of dust in the cloud, and other factors. Some models of the proto-solar disk suggest that the H2O snow line was located about 5 AU from the Sun, not far from Jupiter's current orbit (Cieza et al., 2016). However, hydrous (C-, P- and D-class) asteroids orbit mainly between about 2 and 4 AU (Demeo & Carry, 2014), so either ice formed in that region (by dramatic migration of the snow line) or asteroids were delivered to that region from further out. Proposed mechanisms for delivering ice-bearing asteroids into the asteroid belt include the Nice and the Grand Tack models (Figure 6.9).

  Fig. 6.9 Grand tack and Nice models. No Permission Required Grand tack and Nice models. Red and blue shades indicate the location and spread of S-type and C-,P-,D-class asteroids, respectively.

The Nice model (Gomes et al., 2005; Morbidelli et al., 2005; Tsiganis et al., 2005) describes a scenario whereby the Jovian planets experienced a violent reshuffling event ~3.9 Ga (Figure 6.9). Slow planetary migration was induced in the Jovian planets by gravitational interactions with planetesimals leaving the planetesimal disk. Eventually Jupiter and Saturn crossed a mutual mean motion resonance triggering a reorganization of the solar system. Giant planets moved, existing small body reservoirs were depleted or eliminated, and new reservoirs were created in particular locations.

The Nice model quantitatively explains the orbits of the Jovian planets and Neptune, the orbits of bodies in several different small body reservoirs in the outer solar system (e.g., Trojans of Jupiter), the population characteristics of the Kuiper belt and scattered disk (Levison et al., 2008), the irregular satellites of the giant planets, and the late heavy bombardment on the terrestrial planets ~3.9 Ga (Gomes et al., 2005; Morbidelli et al., 2005; Tsiganis et al., 2005).

One issue with the Nice model is that it predicts transported Kuiper Belt objects (KBOs; things looking like D-class asteroids) should predominate in the outer asteroid belt, but we know only about 10% of the outer main asteroid belt actually look like D-class objects (Bottke et al., 2008). However, based upon collisional modeling, (Bottke et al., 2008) argue that more than 90% of the objects captured in the outer main belt could have been eliminated by impacts if they had been weakly-indurated objects. These disrupted objects should have left behind pieces in the ancient regoliths of other, presumably stronger asteroids. Thus, a derived prediction of the Nice model is that ancient regolith samples (regolith-bearing meteorites) should contain fragments of collisionally-destroyed KBOs. In fact, KBO pieces might be expected to be present in most ancient regolith-bearing meteorites.

Building on the Nice Model, and originally developed to explain the small mass of Mars, the Grand Tack Model proposes that after drifting in to ~1.5 AU, Jupiter reversed course due to capturing Saturn in an orbital resonance, eventually halting near its current orbit at 5.2 AU (Brasser et al., 2016; Walsh et al., 2011). In this model, Jupiter twice crossed the asteroid belt, scattering asteroids first outward and later inward. This mixed initially outer belt C-, P-, and D-class asteroids among the more inner belt S-class asteroids. The resulting asteroid belt has a small mass, a wide range of inclinations and eccentricities, and a population originating from both inside and outside Jupiter's original orbit. The Grand Tack model therefore predicts that bodies (principally C-, P- and D-class asteroids and cometary bodies) formed beyond Jupiter, the CO2 and H2O snow lines, and were later introduced into the asteroid belt. The discovery of a CO2-bearing aqueous fluid inclusion in a CM2 chondrite calcite crystal (Tsuchiyama et al., 2021) supports this hypothesis.

Atmospheric entry heating

Meteoroids are cold soaked in space for thousands to millions of years. In their plunge through the Earth’s atmosphere, heating occurs for only a few seconds and so is experienced only by the outer skin of the body to a depth of only a few mm, as determined by petrographic study (Ramdohr, 1967). In this “fusion crust”, hydrous phases are melted or dehydrated, so volatiles are lost to Earth’s atmosphere. However, below this fusion crust heating was insignificant. Studies of extraterrestrial hydrous phases and organics are always careful to avoid the fusion crust. Of course, problems arise in the study of IDPs and micrometeorites which are more thoroughly heated by atmospheric entry. The influences of atmospheric entry heating are further discussed in Section "Atmospheric entry" of this chapter.

Terrestrial modification of meteorites

Atmospheric entry

The delivery of water and organics onto the Earth’s surface is strongly influenced by their capability of surviving high temperatures during atmospheric deceleration and impacts to the terrestrial surface. The level of atmospheric entry heating depends on the body’s entry angle, velocity, composition, and mass. Asteroids that are larger than 100 m in radius cannot be efficiently aerobraked by the 1-bar atmosphere of the Earth (Chyba et al., 1990), and thus the chances of survival of organics are slim at an entry velocity of over 10 km/s. Therefore, the successful exogenous delivery of organic molecules requires meteorites that are large enough to avoid complete ablation, vaporization or severe heating, and materials that are small enough to be decelerated by the atmosphere, such as MMs and IDPs (Love & Brownlee, 1993). Nevertheless, small particles are more effective in delivering organic carbon to Earth (3.2 × 105 kg/year of organic carbon) as compared to meteorites (7.6 kg/year of organic carbon) (Anders, 1989).

Despite the potential of MMs/IDPs for delivering carbonaceous material to Earth, they are typically frictionally heated at elevated temperatures (400–1200 °C) for several seconds during atmospheric passage (Flynn, 1989; Greshake et al., 1998; Love & Brownlee, 1991; Rietmeijer, 1996, 2004; Sandford & Bradley, 1989) (Table 6.3). Biomolecules such as amino acids could decompose at these temperatures because they begin to pyrolyze at 400 °C (Basiuk & Douda, 1999), and above this temperature at around 500 °C, amino acids can only survive for 1s (Chyba et al., 1990). On the contrary, as the pyrolytic temperatures experienced by meteorites extend to only less than 1 mm depth (which form the dark meteorite fusion crust), organic compounds from the interior of a meteorite is efficiently protected from heating (Sears, 1975).

  Table 6.3 Sizes and thermal history of extraterrestrial objects subjected to atmospheric entry heating.

Sizes and thermal history of extraterrestrial objects subjected to atmospheric entry heating. Modified from Basiuk, V. A., et al, 1999.

|  |  |  |
| --- | --- | --- |
|  Solar system small bodies  |  Size range (m)  |  Atmospheric entry heating  |
| Dust |  |  | <10−6  | No heating |  |
|  |  | 10−6−10−5  | Full depth heating | <500 °C |
| MM/IDP |  | 10−5−10−3  | Up to total evaporation |
| Meteorite |  | 10−3−10 | Surface heating to <1mm depth |
| Asteroid |  | >10 | Explosive impacts up to >10,000 °C |
| Comet |  | >10 | Airburst: moderate heating |

Water and some organic compounds (e.g. amino acids, ketone, PAHs which have vapor pressures at temperature above 150 °C) can survive atmospheric passage via sublimation into the Earth’s atmosphere, by vaporization off the surface of MMs/IDPs before they are melted (Basiuk & Douda, 1999; Basiuk & Navarro-González, 1998; Matrajt et al., 2006). The porosity of MMs/IDPs can also provide an ablative cooling effect (Clark, 1988). Sublimed organics can condense on ice crystals in the cold upper atmosphere behind the meteorite away from the frictional heating at the front, and descend through the mesosphere to reach the Earth’s surface by atmospheric transport and gravitational settling (Glavin & Bada, 2001). In larger meteorites, volatile organic compounds can be vaporized from the peripherical region far from the highest temperature heated zone, and redeposited onto the cold, porous media (e.g. fine-grain matrix) of a meteorite.

The refractory phases cannot vaporize at low temperatures and thus they are subjected to modification/decomposition by the full extent of heating. The heating can alter the isotopic composition and the structure of the organic component by modifying the relative abundance of the aliphatic and aromatic components (Chan et al., 2020). Therefore, heated IDPs often contain a higher abundance of high-mass PAHs (500–700 atomic mass units (amu)) relative to low-mass molecules, as a result of the polymerization/sintering of low-mass compounds by heating (Postberg et al., 2018; P. C. Thomas et al., 2016). Selective decomposition of the thermally labile aliphatic bridges during atmospheric passage could have lowered the isotopic ratio of deuterium to hydrogen (Keller et al., 2004), while the enrichment in the heavy nitrogen isotope can be preserved in heated IDPs as they are hosted in the nitrogen-heterocycles in the heat-resistant high-mass PAHs.

Terrestrial residence

As soon as meteorites reach the Earth’s atmosphere, they are immediately exposed to the terrestrial environment, interact with terrestrial water, air, and biology. The degree of terrestrial weathering of meteorites increases with their terrestrial age (terrestrial residence time of meteorites after they fall to earth), therefore, the longer terrestrial residence times, the higher the degree of terrestrial alteration, environmental conditions being equal.

Meteorites are considered “Falls” if they can be associated with an observed fall event and “Finds” if they cannot be connected to a recorded fall event. Most meteorites (98%) in the terrestrial collection are “Find” meteorites, and most of the Finds were recovered from arid and polar deserts as these environments are the most suitable for the storage of meteorites for long periods of time (at least 50,000 years) (Jull, 2006). The terrestrial age can be estimated by the level of cosmic-ray-produced isotopes (e.g. 14C, 36Cl, 53Mn, 81Kr) in the meteorites (Jull et al., 2000), and there are some chondritic meteorites recovered from Antarctica that have survived for 2 million years (Welten et al., 1997).

Meteorites have been collected from all terrestrial environments: farmland, lake, swamp, concrete, residential backyard, etc. (Figure 6.10). Terrestrial contamination of meteorites is an extremely rapid process (can take place within hours), depending on the environmental conditions of the landing site. Meteorites residing in these organically-enriched environments are contaminated not only on the surfaces, but also in the interior to a depth of a couple of mm to cm (Anders et al., 1964; Glavin et al., 1999; Howe et al., 1965; Kminek et al., 2002), due to porewater fluid infiltrating the stones (Oró et al., 1971).

  Fig. 6.10 Meteorites and their fall sites. No Permission Required Meteorites and their fall sites. (a) The largest fragment of the carbonaceous chondrite Tagish Lake which fell to Earth in 2000, first detected by U.S. Department of Defense (DOD) satellites on 18 January, 2000 . (b) Most fragments of the Tagish Lake meteorite were later collected from the frozen surface (and below) of the lake in northern British Columbia, Canada, in January 2000. (c) Ordinary chondrite Carancas fell to the south of Lake Titicaca in Peru on 15 September, 2007. (d) A 14.2 m diameter crater was formed at the edge of a creek bank by the impact of Carancas. The crater was immediately filled with water as the ground water table was breached. The scales are given by the cubes, measuring 1 cm across.

 During their terrestrial residence, meteorites interact with the fall site environment, and they can accrue terrestrial contamination, as well as losing extraterrestrial organics such as by leaching via exposure to ice meltwater (Glavin et al., 2006). Despite dedicated efforts in the design and execution of contamination control, it is currently impossible to completely eliminate sources of organic contamination (Chan, Franchi, et al., 2020; Dobrică et al., 2012). For examples, a significant portion of the amino acids found in the Allende meteorite are contaminants derived from storage and/or handling (Basile et al., 1984; Belsky & Kaplan, 1970; John R. Cronin & Chang, 1993; Krishnamurthy et al., 1992; Sephton, 2002; Sephton et al., 2004; Yabuta et al., 2007).

Terrestrial weathering and contamination can significantly alter the hydrogen isotope ratios and water contents of meteoritic minerals, even in deserts. Despite the negligible effect of terrestrial contamination on water contents in hydrated meteorites (e.g. meteorites with water content of over 0.2 wt.%) (Engrand et al., 1999), excepting the addition of adsorbed water, a notable difference in hydrogen isotopic ratio has been observed for meteorites (of different residence times) recovered from desert and those collected elsewhere (Ash & Pillinger, 1995; Stephant et al., 2018). These authors studied several fragments broken off from the same meteorite which were exposed to terrestrial weathering in a desert for different durations (0, 1 and 3 years, respectively), and found that while the interiors (>2 mm away from the surface) of these samples remained relatively pristine, the exposed surfaces had absorb terrestrial water, which increased the water abundance, and reduced the hydrogen isotopic compositions after being equilibrated with the deuterium-depleted terrestrial water.

One typical example of potential terrestrial contamination is the Martian meteorite Allan Hills (ALH) 84001, which has caught immense attention and heated debate due to the early announcement of possible bacterial microfossil remain (McKay et al., 1996), however the microfossil was later suggested to be of terrestrial origin, belonging to an Antarctic actinobacteria Actinomycetes (Steele et al., 2000). Another interesting example is the Orgueil meteorite, which is a Fall meteorite that reached the Earth’s surface in 1864 and was recovered to be curated at the Natural History Museum in Montauban, France within 2–4 weeks. Seed capsules and pollen grains were found inside a century later, but were soon recognized as being from a local reed, that were glued into the stone (Anders et al., 1964).

Terrestrial vs extraterrestrial origin

Meteorites and dust particles are delivered to the surface of the Earth, a planet where water, life and biogenic materials are ubiquitous. Unfortunately, the samples will never be cleaner than the tools and containers used to store and handle them, and cannot be completely isolated from the terrestrial environment in which they reside. Rigorous identification of terrestrial organic contaminants relies on the ability to contrast the type, distribution, structures, and isotopic composition of organic compounds between typical terrestrial contaminants and extraterrestrial material.

The extraterrestrial origins of water and organic compounds of meteorites and dust particles can both be established by their compound-specific isotopic compositions, and for organic compounds, three other observations can be made to determine their origins: 1) the presence of organic molecules that are not used and/or synthesized by terrestrial organisms, for example the presence of proteinogenic amino acids (e.g. glycine, leucine, valine) could potentially indicate terrestrial contamination, 2) a distinctive organic content of terrestrial materials collected from the terrestrial fall site (e.g. soil, ice), and 3) a racemic enantiomeric ratio for chiral molecules (see Section "Enantiomeric ratios").

Water

Both hydrogen and oxygen isotopes are informative about the origin of the water in meteorites and dust particles (the calculation of isotopic compositions is given in [Box 6.1](#Box_6_1)). The hydrogen isotopic composition of extraterrestrial water spans a wide range, and the extremes are inherited from a δD value of −865‰ from solar H2 at high temperatures, and a δD value up to +5,400‰ from the ISM/protosolar molecular cloud at low temperatures. D-rich water cannot be produced even in the cooler regions of protoplanetary disks (Cleeves et al., 2014), therefore, the water in the inner solar system is estimated to take lower δD values, and the bulk Earth δD value is estimated to be about −43‰ (C. M. O’D. Alexander et al., 2012).

  Box 6.1 Calculating the isotopic compositions – the δ value

The isotopic composition of a sample is determined relative to a standard material. The standard values in these expressions are the D/H, 13C/12C, 15N/14N, 17O/16O and 18O/16O ratios of the standards.

 Isotopic compositions are reported as δ values, representing the deviation of the measured isotopic ratios from reference terrestrial standards in per mil (‰), where:

 $ δ(‰)=( / R\_{reference} -1)×1000 $

Reference values for isotopic ratios (R) are determined from the reference values of the D/H ratio of standard mean ocean water (SMOW) (Hagemann et al., 1970), the 13C/12C ratio of the PeeDee Belemnite (PDB) standards (Craig, 1953), 15N/14N ratio of the atmosphere nitrogen (Mariotti, 1983), and the 17O/16O and 18O/16O ratios of SMOW (McKeegan & Leshin, 2001). D/HSMOW = 0.00015576; 13C/12CPDB = 0.0112372; 15N/14NAir = 0.003676; 17O/16OSMOW = 0.0003829; 18O/16OSMOW = 0.0020052.

Similarly, oxygen in the water of most extraterrestrial materials exhibits isotopic fractionation. Nearly all terrestrial samples fall along a terrestrial mass fractionation (TF) correlation between δ17O and δ18O with a slope of 0.52, due to the initial homogenization of the terrestrial reservoir (McKeegan & Leshin, 2001). The TF line passes through an origin defined by the terrestrial (SMOW) composition for oxygen. The majority of extraterrestrial materials can be distinguished from terrestrial samples as their oxygen compositions deviate from the TF line (excepting the CI chondrites).

Organic compounds

Isotopic analysis

Extraterrestrial SOM and IOM are typically enriched in D, 13C and 15N (C. M. O’D. Alexander et al., 2007; Aponte et al., 2011; Chan et al., 2021; Cooper & Rios, 2016; Elsila et al., 2012; Krishnamurthy et al., 1992). The heavy isotope enrichments in extraterrestrial organic matter (δD values up to ~30,000% in the IOM of IDPs; (Busemann et al., 2009)) contrast with the lighter isotopic compositions of terrestrial organic matter: δD = –350 to +50 ‰, δ13C = –60 to –25‰, and δ15N = –10 to +20 ‰ (Butterworth et al., 2004; Füri & Marty, 2015) (Figure 6.11).

  Fig. 6.11 Comparison of the δD and δ13C values of terrestrial&amp;nbsp; and extraterrestrial organic matter No Permission Required Comparison of the δD and δ13C values of terrestrial (T) and extraterrestrial organic matter. Data are obtained from Butterworth, et al, 2004; Sephton, Mark A. and Botta, 2005; Alexander, et al, 2007; Martins, et al, 2008; Cooper and Rios, 2016, and references therein.

The enrichments in the heavy isotopes are a signature inherited from their synthetic origin: their organic precursors were synthesized in a very cold (<50K) environment such as the ISM, molecular cloud, and/or outer region of the protoplanetary disk (J.R. Cronin et al., 1995; John R. Cronin & Chang, 1993; Messenger et al., 2003), through interstellar chemical reactions such as gas-phase reactions (e.g. UV driven photo dissociation, radiative association, ion–molecule reactions) and solid-phase reactions that took place when atoms/molecules froze out on the surfaces of interstellar dust grain (Irvine et al., 2000; Tielens, 1983). Molecules observed by radio astronomical observations in the coldest clouds, where isotopic fractionation is efficient, have D/H ratios enriched by 104 times the local D/H (Millar et al., 1989). The fractionation effects are smaller for heavier atoms - the modest enrichments observed in 13C and 15N are consistent with an interstellar cloud origin for the organic precursors inherited from the low value in the change of energy during their ion molecule exchange reactions (John R. Cronin & Chang, 1993; Füri & Marty, 2015).

Compound-specific isotopic analysis (CSIA) of a target organic molecule requires the presence of the compound at an abundance higher than the detection limit of the analytical technique. For example, more than 0.7 nmol of glycine and 0.25 nmol of D-isovaline are required for their CSIA (Elsila et al., 2009; Glavin et al., 2010). Glycine and D-isovaline are present in the Murchison meteorite at ~27 nmol/g and ~9 nmol/g, respectively, meaning that at least 30 mg of Murchison is needed for detecting these amino acids. This becomes particularly challenging for meteorites with low amino acid abundances, such as meteorites that have been exposed to post-hydration thermal metamorphism (e.g. Belgica (B)-7904, Yamato (Y-), Y-82162, Y-980115, Wisconsin Range (WIS) 91600; (Nakamura, 2005; Nakato et al., 2008; Tonui et al., 2014)) (see Section "Challenges in meteoritic analyses and how that can be overcome by modern technology" for further discussions on the challenges in meteoritic analyses). Glycine and D-isovaline are present in Y-980115 at ~0.25 nmol/g and <0.001 nmol/g, respectively (Burton et al., 2014; Chan et al., 2016), meaning that at least 250 g of samples is required for their CSIA analyses (using current techniques). It is extremely difficult for a curation facility to allocate more than 10 g of sample for a single study. Therefore, CSIA is not always feasible, particularly for meteorites that are only available in limited quantities.

Enantiomeric ratios

Life requires selectivity to allow reactions to occur with a high degree of efficiency, and to avoid confusion with other molecules or reactions (Koshland Jr., 2002). The chirality property of amino acids and sugars is an example of this, which serves as a useful tool for determining the biotic vs abiotic origin of these compounds.

RNA, DNA, and polypeptides all form supramolecular right-handed α-helix structures. All living organisms on Earth use exclusively the L-enantiomers of chiral amino acids in proteins, and the D-enantiomers of chiral sugars in polysaccharides and the backbones of RNA and DNA (Bada, 1995). L- means “levo” or left-handed, and D- means “dextro” or right-handed, and the exclusiveness of the “selected” chirality is known as homochirality.

Although proteins are made up of exclusively L-amino acids, D-amino acid enzyme can also fold into a stable structure, and yet, in nature, D-amino acids only occur in short peptides. Proteins made up of a mixture of D- and L-amino acids cannot fold into bioactive protein secondary structures, and thus cannot form efficient enzymes (Skolnick et al., 2019). Therefore, while biotic synthetic pathways yield homochirality, abiotic syntheses of chiral compounds without the presence of chiral catalysts can only yield racemic mixtures (equal amounts of both the L- and D-enantiomers), and this disparity can be used to determine the origin of the detected meteoritic chiral compounds.

Chiral amino acids and sugars have been observed in meteorites. These organic compounds were suggested to be formed by various reactions, and two venerable pathways are the Strecker amino acid synthesis (Strecker, 1850), and the formose sugar synthesis (Butlerow, 1861). These reactions exhibit an interesting synergy between sugars and amino acids, as the presence of either of these molecules determines the enantioselectivity in the synthesis of the other – amino acids catalyze the synthesis of sugars with an enantiomeric excess in the formose reaction, and vice versa in the Strecker hydration of aminonitriles (Blackmond, 2019). As a result, small enantiomeric excesses of the L-enantiomers of amino acids (Glavin & Dworkin, 2009; Pizzarello et al., 2003; Pizzarello & Cronin, 2000) and D-enantiomers of sugars (Cooper & Rios, 2016) have been observed in meteorites, which leads to the hypothesis that extraterrestrial materials may have seeded the earth with a small initial enantiomeric imbalance, of which the ratio was eventually amplified and developed into homochirality.

Challenges in meteoritic analyses and how that can be overcome by modern technology

Valuable information about the origin and evolution of meteorite parent bodies (mainly asteroids) can be obtained from mineralogical and petrological observations, and a micro-analysis of the physical properties of meteorites (Fujiya et al., 2020). It is a perpetual problem to balance the mass requirements for each measurement against the typically small mass available for the analyses. For this reason, it is necessary to make sequential measurements beginning with non-destructive and moving gradually through increasingly destructive analyses (M. E. Zolensky et al., 2000).

Mineralogy and petrology

Sample primitiveness and degree of aqueous alteration can usually be evaluated based on a meteorite's matrix mineralogy (Noguchi et al., 2017). The mineralogy and petrology of the samples can be compared to remote sensing observations of asteroid surfaces (e.g., the presence/absence of absorption at ~0.7 and ~2.7 µm indicative of hydrated minerals and at ~3.4 µm for macromolecular organic solids) (Hiroi et al., 1993; Rivkin et al., 2002). X-ray diffraction (XRD) analyses can be combined with X-ray microtomography observations, and successive 3D-computed tomography (CT) images can be constructed to reveal the internal three-dimensional structures and identify minerals enclosed within the grains (Tsuchiyama et al., 2021, 2011) (Figure 6.12). These analyses use small grains, 10-50 µm, to acquire basic and precise information.

  Fig. 6.12 Potential fluid inclusion located by µ-XRCT in the Sutter’s Mill meteorite. No Permission Required Potential fluid inclusion located by µ-XRCT in the Sutter’s Mill meteorite. The fluid inclusion is hosted in calcite with a daughter mineral that crystalized in the aqueous solution.

Typical sample preparation methods for organic analyses

If micro-scale organic aggregates are included, a portion of them can be analyzed by micro-Raman spectroscopy to understand the maturation and graphitization of the carbonaceous matter. Other portions of the organic-rich area can be analyzed by NanoSIMS for understanding the local enrichments in 2H (D), 13C, and 15N. Ultrathin sections of organic-rich samples, especially, embedded in resin or sulfur can be produced by ultra-microtomy or Focused Ion Beam (FIB) techniques (T. Noguchi et al., 2011). The embedding material is tailored to the sample and proposed analyses - sulfur is used for organic analyses and epoxy for inorganic analyses - analyzed grains can be handled without interaction with atmospheric oxygen or water vapor during embedding, sectioning, and transportation to the instrument. These ultrathin sections can be observed using Transmission Electron Microscopy (TEM) (T. Noguchi et al., 2017). Multivalent elements such as Ti, V, Cr, and Fe can be investigated for their valence states, or N and C to characterize the molecular structure of organic matter by X-ray Absorption Fine Structure (XAFS) (Sutton et al., 2020) or X-ray absorption Near-Edge Spectroscopy (XANES) (Fujiya et al., 2020).  The ultrathin sections can then be analyzed using a high spatial resolution Secondary Ion Mass Spectrometry (SIMS) (i.e. NanoSIMS) for further information about their isotopic compositions (see below). Further characterization of the samples can be performed using an Electron Probe Micro Analyzer (EPMA) and Electron Back Scattered Diffraction (EBSD) (Fujiya et al., 2020). Before any bulk analyses one must check for terrestrial contamination. For example, one can perform XANES analyses for Fe to monitor a possible change in the Fe valence states resulting from terrestrial alteration (Monkawa et al., 2006).

Isotopic analysis

Non-mass dependent isotopic compositions are a powerful tool to characterize the source materials from which the samples formed and possibly their locations. Different meteorite groups, each of which is generally considered to have derived from distinct types of asteroids, show unique isotopic signatures for these elements (Clayton, 1993; Schiller et al., 2018). Especially, the differences between carbonaceous chondrites (CCs) and non-carbonaceous chondrites (NCs) is pronounced in plots of neutron-rich isotopes of the elements O, Ti, Cr, and Mo, most commonly a plot of Δ17O against ε54Cr or ε50Ti values (Δ17O represents a deviation from the terrestrial fractionation line defined by Δ17O = δ17O - 0.52 × δ18O; ε54Cr and ε50Ti denote parts per 10,000 deviations of 54Cr/52Cr and 50Ti/48Ti ratios, respectively, from terrestrial standard values) (Kruijer et al., 2020; Trinquier et al., 2009; Warren, 2011). These measurements can be made by SIMS, Laser-Ablation-Inductively-Coupled MS (LA-ICP-MS) or Thermal Ionization Mass Spectrometry (TIMS) on µg sized samples with sub-permil resolution (Nakashima et al., 2013; Yurimoto et al., 2011).

Although interpretation of the differences revealed by these diagrams is still developing, it is currently believed that they may be used to discriminate between inner (NC) and outer (CC) solar system origins, and possibly provide a crude measure of formation distance from the sun (still hypothetical) (Goodrich et al., 2021). Bulk to microscale D, 13C, 15N, and heavy noble gas contents are useful for understanding origins because their enrichments are regarded as requiring a cold environment such as the outer solar system or interstellar space discussed above (Charnley & Rodgers, 2002; Kebukawa et al., 2020; Marty, 2012). SIMS and LA-ICP-MS techniques are used to obtain chemical and isotopic compositions in µg sized samples with a spatial resolution of a few to tens of micrometers (Joy et al., 2006). Krypton- and Xe-isotope analyses require larger sample amounts, on the order of ~1 mg, comparable to the sample amounts for O-, Ti-, and Cr- isotope analyses.

Compound-specific separation and characterization

More than 50,000 different organic compounds have been detected in carbonaceous chondrites (Schmitt-Kopplin et al., 2010). Molecular, chemical, and isotopic compositions are especially useful in constraining the low-temperature chemical reactions in the early solar system including aqueous reactions in asteroids (C. M. O’D. Alexander et al., 2007; Kebukawa et al., 2020). Compositions of IOM (e.g., C/N and C/H ratios and 15N values) and SOM provide information on the type and thermal history of asteroids.

For analysis of specific amino acids, amines, and sugars, chromatographic separation is coupled with several types of mass spectrometry (Glavin et al., 2018; Naraoka & Hashiguchi, 2019). Depending on the organic concentration, typically tens to hundreds of mg of sample is currently required for the analysis of IOM and SOM. SOM (e.g., amino acids, amines, nucleobases, and hydrocarbons) in the sample extracts can be separated by gas chromatography (GC) and liquid chromatography (LC) and/or multi-dimensional separation systems involving GC, resulting in accurate molecular identification. Time of Flight (ToF)-SIMS, Desorption Electrospray Ionization (DESI)-MS, and matrix-assisted laser desorption/ionization (MALDI) can be used to evaluate the synthesis and evolution of asteroid organic matter (Naraoka & Hashiguchi, 2019). Volatile organics may be characterized by Cavity Ring-Down laser absorption Spectroscopy (CRDS) (Tittel et al., 2003). Compound specific isotope compositions of extractable organic compounds can be analyzed with gas chromatography/combustion/isotope ratio mass spectrometry (GC/C/IRMS) (Aponte et al., 2011; Chan et al., 2016; Elsila et al., 2009; Furukawa et al., 2019; Pizzarello & Huang, 2002).

Chronometric Dating

Chronological information obtained from radiometric dating of samples reveals timing and duration of important early solar system processes. The formation ages of undisturbed primary materials can be measured using the 26Al-26Mg systematics (Nakashima et al., 2015). The timing of aqueous alteration can be revealed from 53Mn-53Cr ages of aqueously-formed carbonates and ferroan olivine, provided proper standards are available (not always the case) (Doyle et al., 2015). The timing of thermal and/or shock metamorphism which dehydrate and otherwise devolatilize samples can be constrained by 39Ar-40Ar (40K-40Ar) dating (Bogard, 2011), if µg quantities are present, using a noble gas mass spectrometer. However, required sample amounts are dependent on the K contents of the samples, which can be small.

Sample return space missions

Despite the international efforts in studying meteorites, MMs and IDPs in the last few decades, the origins of the water and organic material they contain are not fully understood, as processes such as alteration on the parent body(ies), space weathering, atmospheric entry heating, and contamination during terrestrial residence have physically and chemically altered these samples and masked their intrinsic characteristics. The pristine state of extraterrestrial materials can be altered by changing their inherent states, losing sample components, or adding extraneous components. Unfortunately, it is impossible to eliminate sources of terrestrial contamination in extraterrestrial samples that are collected on Earth, as established above.

Sample return space missions aim to deliver the most pristine, unaltered samples from other celestial objects directly to Earth’s curation facilities with strict isolation conditions. The missions provide the most pristine extraterrestrial samples to be analyzed by a diverse array of state-of-the-art instrumentation only available in laboratories (never in spacecraft, due to limitations of payload weight and size). Determination of the “right” solar system samples, and how to capture, return, curate and perform organic analysis on them is no simple matter. So far, there have been three completed sample return missions with an explicit mandate to collect organic matter: the NASA Stardust mission to comet 81P/Wild 2; the JAXA Hayabusa mission to asteroid 25143 Itokawa and Hayabusa2 mission to asteroid 162173 Ryugu (Figure 6.13). Samples from one more asteroid, 101955 Bennu, will be returned by NASA OSIRIS-REx spacecraft in 2023.

  Fig. 6.13 Sample return missions and the total weight of the returned samples Modified from Chan, Q. H. S., Stroud, R., Martins, Z., & Yabuta, H. (2020). Concerns of Organic Contamination for Sample Return Space Missions. Space Science Reviews, 216(4). https://doi.org/10.1007/s11214-020-00678-7 Sample return missions and the total weight (or target weight) of the returned samples. Missions with an explicit mandate to collect organic matter are shown as yellow symbols. Minimum target total weights of returned samples are shown in the figure for Hayabusa2 (target: 5.4 g) and OSIRIS-Rex (target: 0.06–2 kg).

Previous missions

Organic matter was identified in Stardust samples that chemically and structurally resembled the organic matter previously analyzed in carbonaceous chondrites, and in anhydrous chondritic IDPs thought to have a cometary origin (McKeegan et al., 2006; Sandford et al., 2006). Some of this material was found to have 15N and/or D-rich isotope compositions indicative of formation in a cold environments discussed above (Aikawa & Herbst, 2001; Dartois et al., 2005; Messenger et al., 2003; Terzieva & Herbst, 2000). However, due to the small mass of the captured cometary material (Figure 6.14), the SOM that were detected to be indigenous to the target Comet Wild 2 were amines (Burton et al., 2012; Chan et al., 2018; D.P. Glavin et al., 2010; Kvenvolden et al., 1970; Martins et al., 2007) and the amino acid glycine, which was present at an abundance barely sufficient for CSIA (Elsila et al., 2009). As all Stardust particles were captured by high velocity impact, no laboratory measurement of cometary ice was possible (Conel M. O’D. Alexander et al., 2018).

  Fig. 6.14 Stardust mission sample collector and aerogel capture cell. From Allen, C., Allton, J., Lofgren, G., Righter, K., & Zolensky, M. (2011). Curating NASA’s extraterrestrial samples—Past, present, and future. Geochemistry, 71(1), 1–20. https://doi.org/10.1016/j.chemer.2010.12.003 (a) Stardust collector mounted for microscopy and sample preparation in the Stardust Curation Laboratory of NASA’s Johnson Space Center. (b) Cross-sectional view of the aerogel capture cells sandwiched between two opposing wedges. The tray wall is wrapped with aluminum foil.

Studies of Hayabusa samples indicate that asteroid Itokawa is water-rich (up to >900 ppm of water as hydroxyl group in nominally anhydrous minerals) (Chan et al., 2021; Jin & Bose, 2019)compared to most inner solar system bodies (e.g., 250‒350 ppm for L and LL chondrites based on laboratory measurements) (Jarosewich, 1990). However, organic analyses have been carried out on fewer than ten of the returned Itokawa particles (Chan et al., 2021; Ito et al., 2014; Naraoka et al., 2012; M. Uesugi et al., 2019; Masayuki Uesugi et al., 2014; Yabuta et al., 2014). An extraterrestrial origin has been established recently for the Itokawa organic matter via hydrogen, carbon, and nitrogen isotopic analyses by (Chan et al., 2021). The detected organics was suggested to be derived from high temperature (up to 700 °C) surface-catalyzed reactions (e.g. Fischer Tropsch- or Haber Bosch-type gas-grain reactions) (Figure 6.6) that took place in-situ on Itokawa, as well as by late-stage delivery via infall of primitive carbonaceous chondrites and IDPs.

Current missions

Two asteroid sample return missions have just completed or are in progress, both targeting carbon-rich asteroids. The Hayabusa2 mission, which successfully returned to Earth in December 2020, brought back 5.4 g of regolith dust to cm-sized chips from C-type asteroid Ryugu (Watanabe et al., 2017; Yada et al., 2021). The OSIRIS-REx mission plans to return samples from B-type asteroid Bennu in 2023 (Lauretta et al., 2017). Both asteroids are thought to be related to carbonaceous chondrites, thus, returned samples from these two targets should provide organic matter for laboratory analyses designed to address questions regarding the diversity of the organic matter delivered to the surface of the early Earth.

Near-infrared (NIR) reflectance observations by the Near-Infrared Spectrometer (NIRS3) and Visible and InfraRed Spectrometer (OVIRS) on board the Hayabusa2 and OSIRIS-REx spacecrafts, respectively, have detected a ~2.7 µm absorption feature on the surfaces of Ryugu and Bennu (Hamilton et al., 2019; Kitazato et al., 2019). The ~2.7 µm absorption feature indicates the presence of structural hydroxyl (OH) ion in hydrous clay minerals, and that both asteroids have experienced aqueous alteration in the past. The homogenous occurrence of the narrow and weak Mg–OH feature (from Mg-rich phyllosilicates) of spectra obtained from Ryugu’s surface suggests partial dehydration and decomposition of the phyllosilicates by heating. On the contrary, the heterogeneity observed for Bennu’s reflectance spectra, and the absence of the Mg–OH feature in them, indicate a non-Mg endmember (Fe-bearing) phyllosilicate composition, suggesting limited heating of Bennu (DellaGiustina et al., 2020; Hamilton et al., 2019). Together, Hayabusa2 and OSIRIS-REx will reveal the primitive water and organic compositions of unheated/briefly-heated carbonaceous asteroids, and how these signatures could have been altered by short-term heating (e.g. solar radiation / impact) and space weathering that are common to all airless solar system bodies.

Other sample return mission concepts

Although the CAESAR mission was not selected under the NASA’s New Frontiers Program, it remains a well-developed comet nucleus sample return mission concept. The first mission that targeted to return up to 200 g of soil from the Martian moon Phobos by 2014 was Russia's Phobos-Grunt mission (Marov et al., 2004), however the mission terminated in 2012 upon a malfunction that stranded the spacecraft in Earth orbit. A JAXA-led sample return from the Martian moons – the Martian Moons eXploration (MMX) mission, planned to launch in 2024, targets to return samples to Earth from Phobos, with a possible return in 2029 (Krüger et al., 2019; Kuramoto et al., 2018; Usui et al., 2020). Analysis of organic matter is also a major motivation behind sample return mission concepts for Mars, Enceladus, and Titan, among others (Barucci et al., 2012; Mattingly & May, 2011; Tsou et al., 2012).

Conclusions

Exogenous delivery of extraterrestrial water and organic matter to the Earth surface has become an unequivocal knowledge built upon decades of careful investigations of meteorites, cosmic dusts and samples from other celestial bodies returned by sample-return space missions. Such primitive and pristine samples, such as carbonaceous chondrites, provide a rare opportunity to investigate the prebiotic chemistry available to the early Earth, of which any similar information would have been erased by the active terrestrial processes. Meteorites host organic compounds that belong to the same broad groups of compounds found in biomolecules (e.g., amino acids, nucleobases, polyols, carboxylic acids). However, the exact compounds (proteinogenic [e.g. glycine] vs non-proteinogenic amino acids [e.g. isovaline]; nucleobases utilized in biology [e.g. adenine] vs those that are terrestrially rare [e.g. 2,6-diaminopurine]), molecular chirality, and isotopic compositions of extraterrestrial material are distinct from those favored by biology on Earth. Further investigations will equip us with a better capability to evaluate the synthetic origins of extraterrestrial water and organic matter, how they evolved through time on asteroidal parent bodies, and their roles in the origin of life on Earth.

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