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Organic Matter in Cosmic Dust

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Abstract

Organics are observed to be a significant component of cosmic dust in nearly all environments where dust is observed. In many cases only remote telescope observations of these materials are obtainable and our knowledge of the nature of these materials is very basic. However, it is possible to obtain actual samples of extraterrestrial dust in the Earth’s stratosphere, in Antarctic ice and snow, in near-Earth orbit, and via spacecraft missions to asteroids and comets. It is clear that cosmic dust contains a diverse population of organic materials that owe their origins to a variety of chemical processes occurring in many different environments. The presence of isotopic enrichments of D and ¹⁵N suggests that many of these organic materials have an interstellar/protosolar heritage. The study of these samples is of considerable importance since they are the best preserved materials of the early Solar System available.

Keywords

Cosmic Dust; Organics; Carbonaceous Matter; Hydrocarbons; Microanalysis

INTRODUCTION

Dust is seen in a wide variety of astrophysical environments ranging from stellar outflows, the diffuse interstellar medium, dense interstellar clouds, and disks surrounding forming stars. Closer to home, our own Sun is surrounded by a cloud of dust, the Zodiacal Dust Cloud, formed by dust from both comets and asteroids. Our knowledge of the nature of the dust in these environments is based on telescopic observations, spacecraft *in situ* observations, and laboratory studies of collected dust particles. The dust in all these environments consists of a number of different components, the primary ones being minerals and organic materials but in very cold environments condensed gases in the form of mixed molecular ices may also be present.

We provide a brief discussion of organics outside our Solar System, largely for context, and then restrict ourselves to discussion of laboratory studies of organics in Interplanetary Dust Particles (IDPs) collected in the Earth’s stratosphere, unmelted micrometeorites (MMs), and

cometary particles returned by the *Stardust* mission and studied *in situ* by the *Rosetta* mission.

The study of organics in extraterrestrial materials is an inherently difficult activity. Most extraterrestrial materials contain organics at the few percent level that are finely dispersed and amorphous. The organic populations present are often diverse, so that no individual compound or class of compounds is very abundant (e.g., the abundances of amino acids in meteorites are typically reported in ppm). These difficulties are exacerbated for organics in cosmic dust where the samples are small - identifying a multitude of different organic compounds in the few percents of organic material in a nanogram sized dust particle is not a trivial matter! As a result, our knowledge of organics in cosmic dust is often restricted to the determination of overall organic abundance and basic characterization of the classes of compounds present. Finally, since we live on a planet rife with complex organic compounds, the study of extraterrestrial organics requires constantly vigilance for complications associated with possible terrestrial contamination.

ORGANICS IN THE INTERSTELLAR MEDIUM

The study of the organic component of cosmic dust and gases found outside our own Solar System is restricted to telescopic spectroscopic observations. These provide general information about the nature of organics found in stellar outflows, in the diffuse interstellar medium, and in the cold, dense clouds in which new stars and planetary systems form. These observations demonstrate that hydrogenated amorphous carbon, polycyclic aromatic hydrocarbons, and related materials (cf. Allamandola *et al.* 1999; Dartois and Muñoz-Caro 2007) are some of the dominant forms of complex carbon in space. Aliphatic hydrocarbons are also observed in some environments, including in the diffuse interstellar medium (cf. Sandford *et al.* 1991). In the very low temperatures (10–100K) of dense interstellar clouds, all these organic materials are expected to be largely condensed out onto refractory dust particles along with more volatile species like H₂O, CH₃OH, NH₃, CH₄, CO, CO₂, etc. (Boogert *et al.* 2015). Laboratory experiments have demonstrated that energetic processing by UV, electrons, and cosmic ions modify the chemical composition of the ices and produce additional organic molecules, including organic refractory materials that can survive higher temperature and can be found in objects like meteorites, Antarctic MMs, and IDPs (cf. Dworkin *et al.* 2001). Subsequent irradiation of these ‘ice residues’ can then alter the original organics and generate new ones. The organics originally incorporated into our Solar System reflect a long and varied history involving a wide variety of environments and chemical processes. This initial complexity was then compounded by subsequent chemical processing in the protosolar nebula, asteroids, and comets.

STRATOSPHERIC INTERPLANETARY DUST PARTICLES (IDPs)

Dust particles released from asteroids and comets migrate through the Solar System under the influence of both gravitational and non-gravitational forces that can ultimately lead them into collision with the Earth’s atmosphere. Materials that survive deposition into the upper atmosphere drop to the Earth’s surface on timescales of minutes to weeks depending on their masses and densities. It is therefore possible to both collect dust sized particles as they settle

through the stratosphere using specialized aircraft or balloon-borne instruments (Rietmeijer *et al.* 2016; see also the articles by D. Brownlee and S. Taylor in this same issue).

Given the extremely small size of extraterrestrial dust collected in the stratosphere (particles typically have masses of ~1 ng), most of what we know about them is based on a handful of micro analytical techniques, including infrared and Raman spectroscopy, C,N,O X-ray Absorption Near Edge Spectroscopy (XANES), Scanning Transmission X-ray Microscopy (STXM), Laser-desorption Laser-ionization Mass Spectrometry (L²MS), Scanning and Transmission Electron Microscopy (SEM and TEM), and Secondary Ion Mass Spectrometry (SIMS and NanoSIMS).

Chondritic porous interplanetary dust particles (CP IDPs), considered the least altered samples of the Solar Nebula dust, often contain carbonaceous material and in many cases show significantly higher bulk carbon abundances than do carbonaceous chondrites (e.g. Thomas *et al.* 1993; Figure 1). The carbon in stratospheric IDPs shows a wide range of morphologies, including poorly graphitized carbon, coatings on mineral grains, and discrete ‘nanoglobules’ (e.g. Keller *et al.* 2004; Matrajt *et al.* 2013 and references therein). High-resolution X-ray imaging and spectroscopy on ultramicrotome sections of CP IDPs show a ~100 nm thick coating of organic matter often exists on the surface of individual submicron mineral grains within IDP aggregates. These organic layers may have acted as a ‘glue’ that increased the efficiency of dust particle aggregation in the Solar Nebula (Flynn *et al.* 2013).

Much of the carbonaceous material in IDPs consists of amorphous aromatic materials, but aliphatic moieties and C=O groups are also present (Flynn *et al.* 2013). The population of aromatic species includes a wide variety of polycyclic aromatic hydrocarbons (PAHs) and their alkylated derivatives, some having masses >500 amu (Clemett *et al.* 1993). The PAH population sometimes shows a two-lobed mass distributions with peaks centered near ~250 amu and ~400 amu. It is possible that the higher mass part of the distribution is due to thermal alteration (sintering) produced during atmospheric entry heating.

Isotopic studies show that many IDPs exhibit extreme H and N isotopic heterogeneity on the sub- μ m scale. The most typical variations are excesses in D and ¹⁵N that are generally attributed to organics (e.g. Keller *et al.* 2004; and references therein). The presence of such anomalies demonstrates the materials are not terrestrial contaminants. Furthermore, since D and ¹⁵N enrichments are expected signatures of many types of astrochemistry, particularly chemistry in very cold environments like those found in dense interstellar clouds - the environments where new stars and planetary systems form (cf. Sandford *et al.* 2001) - their presence suggests their carriers (or at least their precursors) have origins in the interstellar medium and/or outer portions of the protosolar disk, i.e., these materials in some ways could predate the Solar System.

ANTARCTIC MICROMETEORITES

Micrometeorites are IDPs collected at the Earth surface, most efficiently on the polar caps. They range in size between 20 μ m and 2000 μ m, the smaller being the more abundant. Antarctic micrometeorites are mostly related to a primitive class of meteorites, the

carbonaceous chondrites (Engrand and Maurette 1998), which represent a few percent of the meteorites collected on Earth. Chondritic micrometeorites contain a few wt% of Insoluble Organic Matter (IOM) that has a primitive and very disorganized structure (Engrand & Maurette 1998).

Micrometeorites collected from blue ice at Cap-Prudhomme in Antarctica contain soluble organic molecules such as PAHs (Clemett *et al.* 1998; Figure 2) and amino acids (Matrajt *et al.* 2004; and references therein).

Ultracarbonaceous micrometeorites (MMs) have been found in the Dome Fuji collection (Nakamura *et al.* 2005) and in the CONCORDIA micrometeorite collection made at Dome C (Fig. 3) (Duprat *et al.* 2010). These particles have carbon contents up to 10 times that of the most C-rich carbonaceous chondrites, i.e., up to 85% organic matter by volume (65 wt% of C) (Dartois *et al.* 2013 and references therein). Such concentrations are comparable with the most C-rich stratospheric IDPs (Thomas *et al.* 1993; Keller *et al.* 2004), and compatible with 'CHON' grains (grains rich in C, H, O. and N) detected in comet Halley by the Giotto and Vega spacecraft.

The organic matter in ultracarbonaceous MMs show extreme D/H ratios, and is significantly enriched in nitrogen compared to meteorites, with nitrogen abundances reaching ~ 20 atomic percent in the organic matter (Duprat *et al.* 2010). The organic material is low in oxygen and CH₂/CH₃. Overall, ultracarbonaceous MMs have many mineralogical, isotopic, and spectroscopic similarities with cometary grains. Their organic matter could have been produced by irradiation of N- and C-containing ices in dense clouds, colder portions of the protosolar nebula, or possibly at the surface of comets.

Recent analyses by combined STXM-μXANES and SEM/TEM have shown the presence of two distinct phases of carbonaceous matter in ultracarbonaceous MMs having different N contents - the N-rich phase (atomic N/C up to 0.2) shows a smooth texture and is devoid of minerals, whereas the N-poor phase is associated to fine-grained minerals (Engrand *et al.* 2015). This suggests that ultracarbonaceous MMs contain organics that formed by different mechanisms and/or in different locations in the protoplanetary disk. The soluble organic content of ultracarbonaceous MMs has yet to be characterized.

COMETARY ORGANICS SEEN BY THE STARDUST AND ROSETTA MISSIONS

Dust samples were collected from Comet 82P/Wild 2 in 2004 when the Stardust spacecraft flew through the comet's coma at a relative velocity of ~6.1 km/sec. The samples were subsequently returned to Earth in 2006 and have since been available for laboratory study.

Stardust had a passive dust impact collector made of aerogel, an extremely low density Si-O based material that contained some residual carbon. Despite the use of low density aerogel, impacting particles suffered varying degrees of damage during impact and, unfortunately, organic materials generally survived impact less well than mineral grains. In some particles much of the original organic material was mobilized and injected into the surrounding

aerogel, as evidenced by infrared spectra of ‘halos’ of -OH, aromatic -CH, aliphatic -CH₂- and -CH₃, and C=O groups seen surrounding the physical tracks (Sandford *et al.* 2006). Halos of organics are not seen around tracks when samples of carbonaceous meteorites are fired into aerogel, suggesting that the halos in the Stardust aerogel are due to organic materials that are more labile than those typically seen in meteorites. Comparisons between the IR spectra of the organics in IDPs and Wild 2 particles with those in the diffuse ISM suggest both Solar System materials have much higher CH₂ to CH₃ ratios than the interstellar organics (Flynn *et al.* 2003). In the case of the Wild 2 particles it is difficult to assess the extent to which this material was altered from its original form in the particle.

The diffuse dispersion of organics in the aerogel is demonstrated by the detections of amines and amino acids with non-terrestrial isotopic ratios (Elsila *et al.* 2009) in samples of non-track-containing aerogel. Whether these compounds were volatilized from impacting particles or simply arrived at the collector in the gas phase is unknown.

Fortunately, there are cases where organics in the particles survived largely intact and unaltered because they were protected during impact by surrounding minerals (Figure 4; Matrajt *et al.* 2013). In these cases, organics can be seen distributed throughout the terminal particle or as individual concentrations within them (Rotundi *et al.* 2014).

Given the small amounts of organics in terminal particles, most of what we know is based on use of the same micro-analytical techniques used to study stratospheric IDPs, including XANES (Sandford *et al.* 2006), infrared and Raman spectroscopy, and SEM (Rotundi *et al.* 2014), L²MS (Clemett *et al.* 2010), and SIMS (McKeegan *et al.* 2006). Many of the cometary organics are similar to those found in other extraterrestrial materials. For example, Raman data suggests that the Stardust samples contain material similar to the IOM that dominates the organics in meteorites (Sandford *et al.* 2006; Rotundi *et al.* 2014). Stardust samples also contain similar three-dimensional organic structures, such as ‘nanoglobules’ (De Gregorio *et al.* 2010) as those found in carbonaceous meteorites and IDPs (Matrajt *et al.* 2013 and references therein).

However, analyses of Stardust particles also show these samples are more diverse than those found in other extraterrestrial materials and contain additional types of organics that are absent or considerably less abundant in meteorites and IDPs. The XANES, infrared, L²MS, and Raman data all indicate that more labile, aromatic-poor materials are also sometimes present. The diversity of organic material present is demonstrated by nanoSIMS measurements, which show that C/N ratios can vary enormously across a single particle (Sandford *et al.* 2006).

Isotopic analyses demonstrate that some of the organics contain isotopic anomalies in the form of enrichments of D and ¹⁵N relative to terrestrial H and N (McKeegan *et al.* 2006). As with the stratospheric IDPs, the presence of such anomalies demonstrates the materials are not contaminants and suggests an interstellar and/or protosolar heritage.

While the European Space Agency’s Rosetta Mission to Comet 67P/Churyumov-Gerasimenko (hereafter 67P) will not return samples to Earth, it is equipped with several instruments designed to study dust in the comet’s coma. These include GIADA (Grain

Impact Analyser and Dust Accumulator), which measures the number, mass, momentum, and velocity distribution of dust particles in the near-comet environment and COSIMA (Cometary Secondary Ion Mass Analyzer), which images dust collected on metallic targets and measures the elemental and isotopic composition of key elements.

GIADA collected compact particles (ranging in size from 0.03 to 1 mm) and fluffy aggregates (ranging in size from 0.2 to 2.5 mm) of sub-micron grains, with bulk density constrained in the range $(1.9 \pm 1.1) \times 10^3 \text{ kg m}^{-3}$ and up to 1 kg m^{-3} , respectively (Rotundi *et al.* 2015). The fluffy aggregates are probably similar to the aggregates collected by COSIMA (Figure 5) and are probably related to cosmic dust particles collected on Earth (Schulz *et al.* 2015).

While no organics have been directly detected in these grains, the Cometary Sampling and Composition experiment on Rosetta's lander Philae did detect a suite of 16 molecules of which all but one (H_2O) contained one or more C atoms (Goesmann *et al.* 2015). Thus, it is likely that organic species are common in the cometary grains from this comet.

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SUMMARY

Cosmic dust particles contain organics in a variety of morphological forms. The populations of organic compounds present in these particles are complex and appear to vary from particle to particle, implying these materials have complex histories. The presence of isotopic enrichments in D and ^{15}N in some of these materials suggest this history extends back to the interstellar dust cloud from which our Solar System formed, i.e., some of these materials may well be older than the Solar System itself. Since these organics are presumably the result of universal processes that occur whenever and wherever new stars and planetary systems form, similar materials likely commonly fall onto planetary surfaces elsewhere. If such materials played an important role in the origin of life on Earth, they are expected to be available to play a similar role on other planets throughout the universe.

Figure 1.

(a) SEM image of a stratospheric IDP; (b) corresponding high magnification images by transmission electronmicroscopy showing that the particle is an aggregate of smaller grains; (c) Compositional map corresponding to the TEM image in (b) (Thomas *et al.* 1993). Note that carbonaceous material, indicated by the light gray regions in (c), is present at the 40–50% level. By comparison, carbonaceous chondrites, meteorites rich in carbon, typically only contain 3–5% carbon.

Figure 2.

Spectra of PAHs detected in Cap Prud'homme Antarctic micrometeorites using the L²MS technique in which an initial laser is used to vaporize material from the sample and a second laser ionizes selective molecular species before they are passed into a mass spectrometer (from Clemett *et al.* 1998). In this case the ionization laser is tuned to give good yields for PAHs. PAHs consist of fused benzene rings and example PAH structures are shown (moving from left to right) for peaks consistent with the PAHs phenanthrene, pyrene, and chrysene.

Figure 3. Secondary (left) and Backscattered (right) electron micrographs of a fragment of an Ultracarbonaceous Antarctic Micrometeorite deposited on carbon tape. All dark grey patches in the right image are organic matter. Brighter particles are silicates and Fe-Ni sulfides.

Figure 4.

Energy-filtered TEM images of a microtome slice of a Stardust particle. (Left) An image obtained using zero-loss filtering to provide contrast improvement. (Right) An image made using energy passbands above and below the 285 eV carbon edge; in such images the presence of carbon is indicated by light areas. The sulfide at the upper right of the images is C free, but regions of C are clearly seen in the fine-grained chondritic material below the sulfide grain (adapted from Matrajt *et al.* 2013).

Figure 5.

Example of an agglomerate dust particle from comet 67P/Churyumov-Gerasimenko collected by COSIMA (Cometary Secondary Ion Mass Analyzer) (from Schulz et al. 2015). The image was obtained using grazing illumination from the right. The length of the shadows indicate that the altitude above the substrate reaches about 100 μm .