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Organic matter in comets and cometary dust

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Summary. Comets are primitive conglomerates of the solar system containing a mixture of frozen gases, refractory grains, and carbonaceous particles rich in biogenic elements. The dramatic display of comets is mostly caused by a cloud of micrometer-sized dust particles that leave the comet nucleus when frozen gases sublimate as they approach the Sun. Analyses of cometary dust captured in the stratosphere together with data obtained from space missions to comets have revealed the presence of a great variety of organic molecules. Since substantial amounts of cometary dust were gently deposited on Earth, their organic content could have played a major role in prebiotic processes prior to the appearance of microorganisms. This review discusses the description and implications for life of the organic content of comets and cometary dust. [**Int Microbiol** 2005; 8(1):5-12]

Key words: comets · cometary dust · interplanetary dust · prebiotic chemistry · chemical evolution · origin of life

Introduction

Laboratory analyses of primitive meteorites and interplanetary dust particles together with astronomical and spacecraft measurements of comets have revealed the presence of complex organic molecules in extraterrestrial samples. Interest in these organic compounds has remained high since their discovery, because of the information that they can supply concerning a key step in the sequence of events leading to the origin of life, namely, the abiotic chemical reactions that generated the primordial soup. Over 650 individual organic molecules have been identified in primitive meteorites, in addition to more complex macromolecular materials with poorly resolved structures. Their occurrence, origin, and relevance for the build-up of biomolecules were discussed extensively in the first part of this review [27].

Of all the debris left over from the formation of the Sun and planets in the solar system, comets contain by far the largest amounts of the hydrogen-, carbon-, sulfur-, and nitrogen-bearing molecules that are needed to support biogenesis. In comets, these elements are considerably more abundant than in primitive meteorites [27]. Because of the small size of comets, which implies the absence of large-scale differentiation and mixing processes, and their site of accretion, far from the Sun, thus avoiding the depletion of volatile molecules, one may expect that both presolar and nebular organic molecules are best preserved in comets.

The universal view that a comet's nucleus is an icy body that contains rocky material is by no means new; it dates back at least to the times of Pierre Laplace, in the early 19th century. However, in 1950, Whipple resurrected the idea and modified it, describing the essential features of comets [45]. Whipple's theory of "dirty snowballs" became the basis of all later work on the chemistry of comets. Water is the dominant ice in comets, and all other ices, such as carbon monoxide, carbon dioxide, methane, ammonia, and methanol, are condensed or trapped within the amorphous water ice as guests, forming what is called a clathrate hydrate. Physical adsorption is particularly effective on amorphous ice at low temperatures, and clathration can occur at temperatures well above the pure condensation points of the volatiles. Comets contain an average of 40-50% water (by mass), 13-16% volatile organics, 13-15% refractory organics, and 22-26% silicates [22].

Dedicated to Prof. Joan Oró (1923-2004). This review is a continuation of that published in *Int. Microbiol.* 7(4):239-248. (See also pp 63-68, this issue.)

Comets are found in several regions around the Sun. One is a region of the solar system beyond Neptune, named the Kuiper belt, which is a reservoir for short-period comets such as Halley. Pluto and its satellite Charon may be regarded as members of the Kuiper belt, and it is even suspected that Triton, the large moon of Neptune, may be a recently captured object [22]. As many as a few billion comets or even asteroids are thought to make up the Kuiper belt. The Hubble space telescope found indirect evidence for 50 to 60 objects in one of its exposures of a small area of the belt. Two of the most outstanding objects from the Kuiper belt are Chiron, whose orbit takes it inside that of Saturn and then out to Uranus, and Pholus, which currently ranges from inside the orbit of Saturn to just beyond Neptune. The short dynamic lifetime of Pholus in its present orbit along with its red color and spectral properties strongly suggest that it recently entered the planetary zone from the Kuiper belt. Pholus is, therefore, a primitive body that has not experienced large-scale sublimation or chemical processing through heating by the Sun [15]. While the orbit of Pholus is comet-like, its dimensions (diameter at least 140 km) are greater than those of a typical comet nucleus (a few tens of kilometers) and are more similar to those of many asteroids. Neither Pholus nor Chiron clearly fall into any of these classifications. Indeed, both classifications have become rather indistinct in recent years, with the realization, from orbital dynamics, that some objects that had been considered asteroids for a long time are inactive comets, and with the detection of cometary activity in some objects designated as asteroids [20]. Comets also exist further out, in the so-called Oort cloud that surrounds the Sun out to a third of the distance to the nearest stars. The Oort cloud may contain trillions of comets that accompany the Sun in its walk around our galaxy.

Dust everywhere

In 1730, Cassini published one of the first scientific papers related to interplanetary dust [10]. He discussed the so-called zodiacal light, a faint brightness of the sky that can be observed upwards from the eastern horizon before sunrise and above the western horizon after sunset. Today we know that zodiacal light is produced by scattering sunlight from dust particles with sizes of 1 to 100 µm that are distributed along a flattened disk centered on the Sun. The most important sources of interplanetary dust are comets (Fig. 1). As the nucleus of a comet approaches the Sun, volatile and icy components sublimate, and dust as well as larger particles are emitted from the nucleus. This is viewed as a long tail, as the dust is accelerated away from the nucleus by radiation pres-

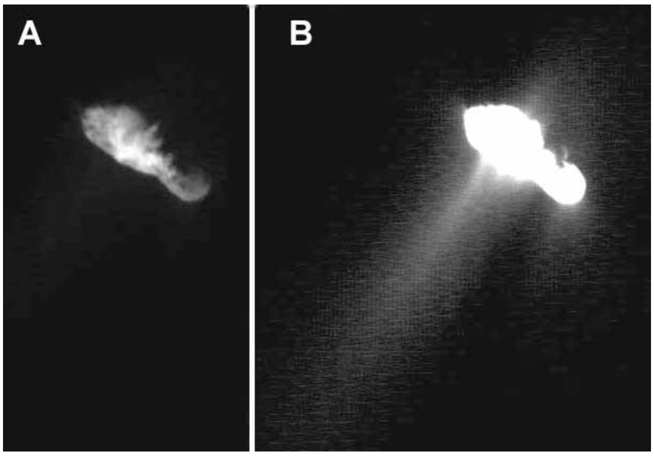

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Fig. 1. Pictures of comet Borrelly taken by the spacecraft Deep Space 1 in September, 2001. (**A**) Nucleus of the comet, which is about 8 km long. (**B**) Enhanced image to reveal dust being ejected from the nucleus of the comet. The main dust jet is directed towards the bottom left of the frame and the direction of the Sun is directly downwards. (Photos courtesy of NASA.)

sure force. This force originates from the interaction of electromagnetic radiation (sunlight) with matter. As an example, the space probes that encountered Halley's comet in 1986 found that the comet was injecting approximately 3,000 kg of dust per second into the interplanetary medium [30]. In fact, the word "comet" derives from the Greek "kometes", meaning long-haired, and refers to a comet's appearance as a tail star; what is seen of a comet with the unaided eye is mostly dust. Other sources of interplanetary dust are asteroids, planetary rings, and interstellar space. Both gravity and radiation pressure from the Sun limit the age of the interplanetary dust particles. Either the particles are decelerated to cause a radial drift into the Sun, where they vaporize, or, if the radiation pressure exceeds gravity, they are blown out of the solar system. Consequently, interplanetary dust is not a remnant of solar system formation but mainly a product of the much more recent evolution of comets and asteroids.

Cometary dust among collected interplanetary dust particles

Interplanetary dust particles (IDPs) have been and still are collected from a wide variety of sites using different techniques. Ocean bottoms, temporary glacial melt lakes in Greenland and Antarctica, space-recoverable rockets and satellites, microcraters on mineral grains from the surfaces of lunar rocks, and the upper atmosphere are some of the environments for IDP recovery [37]. However, collections made in the upper atmosphere are of particular importance since they provide interplanetary dust particles of unquestioned

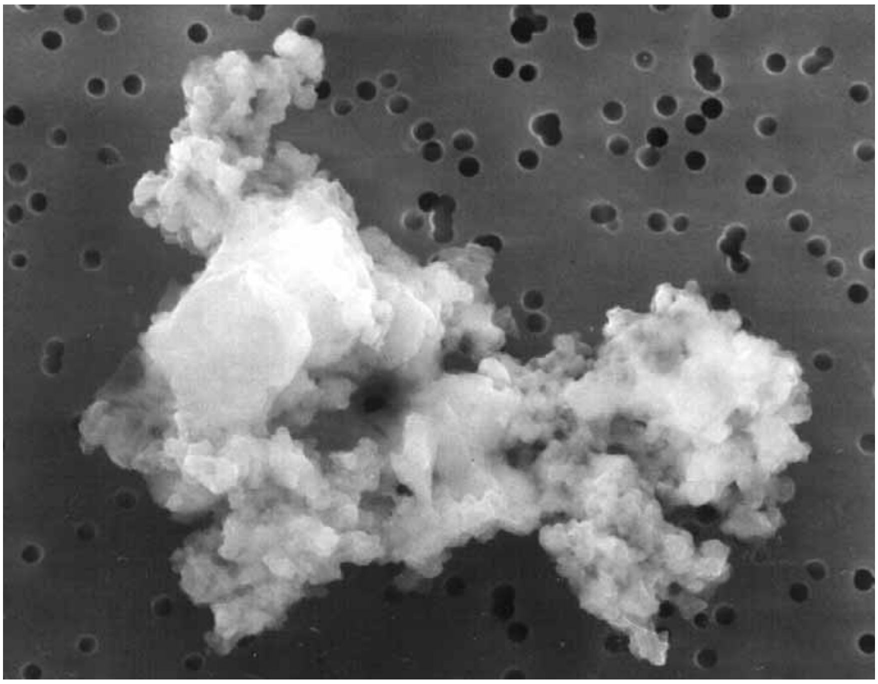

Fig. 2. Scanning electron microscopy image of an interplanetary dust particle collected in the stratosphere. The cluster-of-grapes morphology and composition of this type of particles are consistent with a cometary origin. The particle measures ca. 10 µm. (Photo courtesy of NASA.)

extraterrestrial origin that have not been severely altered by capture processes (Fig. 2). Since 1974 and after extensive work by Donald Brownlee, micrometer-sized IDPs have been collected from the stratosphere with aircrafts flying at altitudes of about 20 km and brought back to terrestrial laboratories for detailed examination [9]. Fortunately, in the stratosphere both natural and anthropogenic particles are rare, so the effective collection of extraterrestrial material is possible, although about one hour of flight is necessary for collecting just one interplanetary dust particle. Common contaminants include aluminum oxide spheres generated by solid-fuel rockets, aluminum- and titanium-rich spacecraft paint, and volcanic ash.

The miraculous aspect of the interaction between interplanetary dust and the atmosphere of the Earth is that deceleration of the IDPs is so gentle that they are slowed without severe heating and mechanical stress. IDPs enter the atmosphere at high velocities, in excess of 11 km/s (for comparison, a cannonball moves at only *ca.* 5 km/s), and slow down at altitudes above 80 km, where the air is very thin. As a result, frictional heat builds up slowly and dust particles radiate the heat without melting [37]. Once decelerated, the particles slowly settle out and can be collected in the stratosphere before they become mixed with the terrestrial particulate material commonly found at lower altitudes.

Several lines of evidence confirm that some of the collected stratospheric dust is extraterrestrial, the most convincing of which are: the presence of implanted solar wind noble gases, including He, Ne and Ar [21], the presence of large deuterium enrichments [47], the discovery of solar-flare tracks in mineral grains within the particles [7], and their chondritic elemental composition (that of carbonaceous chondrites, the most primitive type of meteorites). Chondritic particles, the most common type of IDPs, are roughly subdivided into two major categories, chondritic porous (CP) and chondritic smooth (CS). CS IDPs are relatively compact and their mineralogy is dominated by phyllosilicates; they are similar in many respects to the matrices of CI and CM carbonaceous chondrites [27]. In contrast, CP particles have a characteristic fragile cluster-of-grapes morphology and densities between 0.7 and 2.2 g/cm^3. Most of them are generally anhydrous and consist of a large variety of mineral grains, ranging in size from about 5 nm to a few micrometers. Examination by electron microscopy techniques has revealed that many grains are single minerals, while others are composed of several much smaller crystals embedded in amorphous material. Most of the amorphous material is of a carbonaceous nature (Fig. 3). Amorphous and poorly ordered low-atomic-number phases are abundant in CP IDPs, which occur as clumped masses as well as thin coatings on some grains [28]. An important point to note is that the different minerals in the grains are commonly out of chemical equilibrium with each other, implying that they did not all form at the same time but were instead formed in different environments and then brought together.

The elemental composition of CP IDP grains is similar to that of CI chondrites, but their mineralogy and texture are distinctly different from those of any known meteorite.

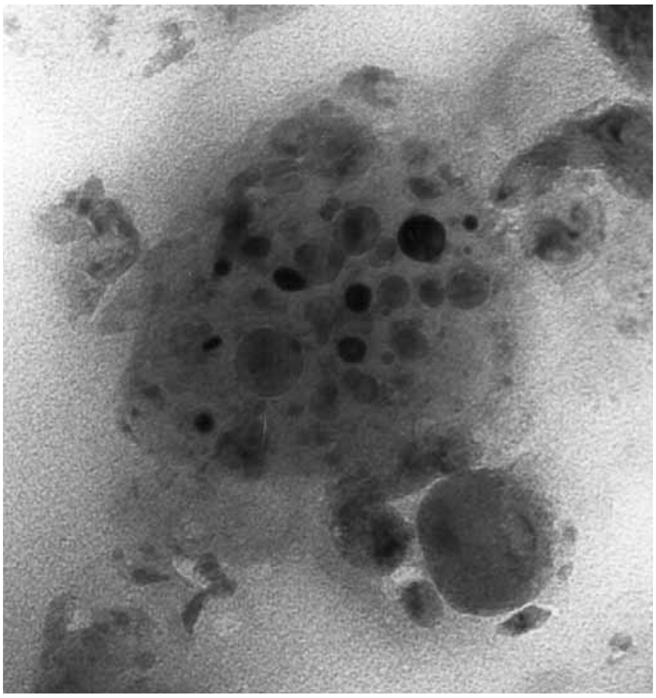

Fig. 3. Transmission electron microscopy image in bright-field mode of a sectioned cometary particle captured in the stratosphere. High-contrast particles are metal grains, which are embedded in a low-contrast matrix of a carbon-rich material. Similar assemblages are obtained in the laboratory after reaction between nebular gases (H_2 + CO + ...) and metal grains with catalytic properties. In the course of these experiments, organic molecules are also synthesized. The aggregate measures ca. 0.1 µm. (Photo courtesy of NASA.)

Taking into account that carbonaceous chondrites significantly deviate from chondritic values on a micrometer scale (attributed to element redistribution by aqueous alteration), CP IDPs represent the least altered material in the solar system. The occurrence of silicate crystals with unusual growth habits formed directly from the vapor phase [8] as well as ε-nickel-iron carbide [6,12], a low-temperature phase that is not present in any other natural system, also suggests that CP IDPs are truly well-preserved ancient material. It is likely that CP IDPs date back to at least the formation of the solar system. In fact, the chemical and physical features of CP IDPs as well as their spectral properties point to a cometary origin for this type of particles [5]. Anhydrous silicates, especially crystalline olivine, has been identified in the spectra of a number of comets [19]. In addition, spectroscopic studies of bright meteors indicate that the relative abundances of Fe, Si, Mg, Ca, Ni, Mn, Ti, and Cr are similar to those of carbonaceous chondrites in most cases [43,44], although the more crucial low-mass, volatile elements cannot be determined.

Some CP IDPs contain small regions of glassy material, the so-called GEMS (glass with embedded metal and sulfide), with appreciable irradiation histories, indicating that they once resided in interstellar space [4]. It is believed that GEMS are interstellar grains that survived in comets, like the interstellar grains recognized in carbonaceous chondrites [27]. Dust particles could act as nuclei for the condensation of gases and organics in dense interstellar clouds as well as in the presolar and solar nebula. They most likely existed in the interstellar medium and survived the collapse in the outer parts of the solar nebula until comet formation.

Biogenic elements in cometary dust

Dust particles released from a comet are not in the same state as they were while residing inside the nucleus. They are devoid of the volatile molecules that may have filled the cavities inside the particles; this is probably why IDPs look like fluffy aggregates. The loose structure of cometary particles is also demonstrated by the low densities of meteors that originated from comets, from which densities as low as 0.1 g/cm^3 have been deduced [11]. Elemental abundances of Halley's comet dust obtained during flybys of the Vega and Giotto missions indicated high abundances of the elements C, H, O, and N in some of the particles, the so-called CHON particles, which exhibited densities in the range 0.1-0.3 g/cm^3 [23]. This was interpreted in terms of organic compounds being present in the cometary dust [24], which appeared to be a mixture of CHON-rich and silicate-rich material present in highly variable proportions. The evidence for carbonaceous material was, in fact, one of the major results of comet Halley's exploration.

The average carbon content of IDPs collected in the stratosphere of the Earth has been measured as 10–12 % [38,41], which is about 2.5–3 times the carbon content of primitive meteorites. Carbon is widespread throughout most IDPs. The small masses of interplanetary dust particles (about 10^{-8} g) have, thus far, precluded quantitative determination of the fraction of this carbon that is present in organic molecules. However, several polycyclic aromatic hydrocarbons (PAHs) and their alkylated derivatives were detected in two IDPs [14]. Also, Raman spectra of individual CP IDPs is dominated by features characteristic of disordered carbonaceous materials rich in aromatic molecular units [2].

Similarly, as has been observed in carbonaceous chondrites [27], some of the CP IDPs contain large bulk deuterium enhancements. Deuterium enrichment have been found to correlate with carbon concentration, suggesting that the D-rich carrier phase is a hydrocarbon [47]. Two major processes could explain the observed deuterium enrichment, both of which operate in the interstellar medium. Ion-molecule reactions at low temperatures in dense molecular clouds yield deuterium-rich simple molecules, which may undergo subsequent processing to complex organic molecules through incorporation into icy grain mantles. Alternatively, selective photodissociation of C-H over C-D bonds in aromatic hydrocarbons may result in D-rich organic molecules [46].

Comets probably originated by accretion of interstellar grains [32,33], and are thus considered to be the least evolved bodies in the solar system. However, interstellar grains were exposed to cosmic and UV radiation prior to accretion and, much later, after the formation of the cometary nucleus, experienced secondary alteration events for 4.6 billion years. For instance, interstellar grains could have been partially vaporized in the nebula and then later recondensed or mixed with processed nebular ices [26,36]. As a consequence, comets probably contain pristine organic molecules as well as more recent complex organic matter.

The organic component in comets

Interplanetary dust particles of cometary origin (CP IDPs) provide insight into the nature of the solid component of comets. However, our knowledge of the nature of the volatile component of the cometary nucleus is based entirely on spectroscopic observations of processes that take place when cometary nuclei interact with solar radiation. As the nucleus approaches the Sun, ices from its surface sublimate. The released gases form a wrapper, called a coma, that is continually lost to the interplanetary medium. Solar ultraviolet radiation dissociates and subsequently ionizes the molecules in the coma, which are, in addition, modified by fast, complex ion-molecule reactions. The ions interact with the solar wind and are blown away into the ion tail [22]. This complicated chain of reactions makes it difficult to reconstruct the nature and abundance of the unobserved parent molecules in the nucleus from the daughter molecular species visible in both the coma and the ion tail. Thus, the precise nature of the organic component in comets remains unknown.

Improvements in spectroscopic techniques have allowed remote detection of volatile organic molecules outgassed from cometary nuclei. The recent close approach to Earth of the comets Hyakutake and Hale-Bopp in 1996 and 1997, respectively, resulted in the first-time identification of the organic species C_2H_2, CH_3CN, HC_3N, NH_2CHO, HCOOH, and $HCOOCH_3$ [3], in addition to other molecules already reported, such as CH_4, C_2H_6, CH_3OH and HCHO. The prebiotic relevance of all these organic molecules is widely recognized [35]. The molecules that have been observed spectroscopically from the Earth in comets are listed in Table 1.

Spacecraft encounters with comet Halley revealed that the nucleus of the comet is heterogeneous and consists of a mixture of brighter and darker materials. The bright component has been interpreted as ices, while the darker one appears to be assemblages of refractory organic and mineral components [22]. Pholus is rich in complex carbon compounds, as deduced from its spectral data [15] and red color. Small, light hydrocarbons, such as methanol and/or its photolytic products, along with H_2O ice occur on its exposed surface. Natural solid-oil bitumens and other complex hydrocarbon materials, such as coal, appear to be spectral analogues for cometary refractory organics [31]. They show color similarities to the surfaces of cometary nuclei, as well as compositional and structural resemblance to organic components of carbonaceous chondrites [27]. These dark solids are composed of a variety of organic compounds, most of which are aromatic and aliphatic hydrocarbons. Comets progressively lose their volatile components as they become older and, at the same time, the organic material on their surfaces gradually becomes more carbonized. In other words, the surfaces of

Table 1. Organic compounds in comets*

Molecule	Relative abundance
H_2O	100
CO	23
CO_2	6
CH_4	0.6
C_2H_2	0.1
C_2H_6	0.3
CH_3OH	2.4
H_2CO	1.1
HCOOH	0.1
CH_3CHO	0.02
$HCOOCH_3$	0.08
NH_2CHO	0.02
NH_3	0.7
HCN	0.25
HNC	0.04
HNCO	0.1
CH_3CN	0.02
HC_3N	0.02
H_2S	1.5
CS_2	0.2
CS	0.2
SO_2	0.2
SO	0.3
OCS	0.4
H_2CS	0.02
NS	0.02

*Updated from Bockelée-Morvan et al. [3].

old comets become depleted in hydrogen-rich aliphatic compounds and richer in condensed aromatic polymers. Chiron, a very dark object, is an example of a heavily processed comet nucleus.

A wide array of organic compounds have been produced by irradiating cometary ice-analogues in the laboratory [1,29,40,42]. When the resulting laboratory ices are warmed up, many of the parent species as well as new photoproducts sublimate out of the ice. At this point, moderately complex organic molecules, such as alcohols, amides, amines, ketones, and nitriles, are formed. In addition, species with greater complexity are produced, including polyoxymethylene-related species ($[\text{-}CH_2O\text{-}]_n$) and hexamethylenetetramine ($C_6H_{12}N_4$). On Earth, many of these species are of great biological importance. When hexamethylenetetramine is hydrolyzed in acid, amino acids are spontaneously produced. More intriguingly, when organic mixtures left over from warming laboratory ices to room temperature are placed in water, insoluble lipid-like droplets form that show self-organizing, membrane-forming behavior [1].

Traveling around the Sun

With the exception of the Earth, Terrestrial planets are poor in organic material because they formed in the inner, hotter regions of the solar nebula. In contrast, as the distance from the Sun increases, the amount of organic matter appears to increase. Asteroids begin to show signs of significant organics, and these are abundant in comets. Organic matter may be common in the moons of giant planets. This is one of the paradoxes of the origins of life: organic molecules thought to be the building blocks of the first microorganisms are found in the outer solar system, whereas the environments conductive to molecular evolution and the growth of life are found in the inner solar system. One possible answer would be that biogenic organic molecules which originated in the outer solar system were transported to the inner solar system. This is the key role that comets and meteorites may have played and is the basis for the current interest in comets and meteorites in the study of the origin of life. In addition, comets may have played a major role in transporting water, essential for the appearance of microorganisms.

According to some estimates, the contemporary influx of meteoritic material—meteorites, comets and interplanetary dust—into the Earth is about 10^5 kg/day [35]. Taken into account that collisions of comets and meteorites with planetary surfaces could result in the decomposition or alteration of their organic content to a considerable extent, interplanetary dust, which is not subjected to significant heating during atmospheric deceleration, is believed to be the best delivery vehicle for organic matter of space origin to the Earth. There are only a few IDPs in each cubic meter of space, but the Earth sweeps up around 10^7 kg of this material each year as it travels around the Sun. About 80% of the total mass accreted by the Earth corresponds to particles weighing between 10^{-7} and 10^{-3} grams [17]. Concerning the survival of organic compounds, Chyba et al. calculated that most of the organic material in planetary materials can survive temperatures up to 850 K for about 1 s [13], and that a typical IDP spends only a few seconds within 100 K of its peak temperature on atmospheric entry. Textural zoning attributable to temperature gradients observed in 50- to 100-µm diameter micrometeorites recovered from terrestrial polar ices suggests that a mechanism exists to allow the interior to remain cool. It has been proposed [16,35] that, during the first 0.5 Myr of the planet's history, more than 10^{12} kg of fine dust preserving the organic compounds of IDPs may have been captured by the Earth.

With the exception of Halley's cometary grains, it has not yet been possible to measure directly the chemical composition of cometary material. As explained above, our current knowledge about cometary volatiles comes largely from spectroscopic observations taken from the Earth, at distances of millions of kilometers. Kissel and Krueger concluded, through mass spectrometric measurements of Halley's dust, that it contains linear and cyclic, saturated and unsaturated hydrocarbons, nitriles, amines, imines, nitrogen-bearing heterocycles, such as pyrroles, pyrimidines, pyridines, purines, etc. [25]. They suggested that the dust particles consisted of a mineral core with an organic mantle, similar in many respects to the laboratory models proposed by Greenberg [18]. Adenine, a significant organic molecule for prebiotic chemistry identified among Halley's purines, could be synthesized in comets, as Oró showed in 1960, from the reaction of hydrogen cyanide in an aqueous ammonia mixture [34, see also pp 63-68, this issue]. Also, Schwartz et al. have demonstrated adenine synthesis from HCN alone in ice [39]. Virtually all of the bases for RNA or DNA can be generated readily from cometary molecules. For this and other achievements, the International Astronomical Union has recently renamed asteroid 1999 XL36 (discovered by the Observatory of Mallorca in 1999) as asteroid 25472 Joanoró.

Did comets participate in the origin of life? The question remains unanswered, but in the next decade direct sampling of the comets Wild 2 and Churyumov-Gerasimenko, as part of the Stardust (NASA) and Rosetta (ESA) missions, initiated in 1999 and 2004, respectively, will provide new information. The implications of these missions and others already planned are huge. We may obtain direct evidence that interplanetary dust particles are well-preserved cometary material. Eventually, we may be able to carefully examine the organic content of comets in the laboratory, as has been done with primitive meteorites for decades. The ability to examine cometary material in a laboratory setting will make these missions invaluable. It could confirm and put into context previous laboratory findings that complex organic molecules may have formed in space and seeded the early Earth. Also, on July 4, 2005, the Deep Impact spacecraft will arrive at comet Tempel 1 to impact it with a 370-kg mass. On impact, a deep crater will be produced, allowing fresh material to be studied using cameras and spectrometer records taken from the spacecraft. The effects of the collision with the comet will also be observable from Earth. We may learn about the structure of a comet's interior and how it differs from its surface.

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Materia orgánica en cometas y polvo cometario

Resumen. Los cometas son conglomerados primitivos del sistema solar que contienen una mezcla de gases helados, granos refractarios y partículas carbonáceas ricas en elementos biogénicos. La apariencia espectacular de los cometas se debe principalmente a una nube de partículas micrométricas de polvo que abandonan el núcleo del cometa a medida que los gases helados se subliman al acercarse al Sol. Tanto el análisis de polvo cometario capturado en la estratosfera como las misiones espaciales a cometas han demostrado la presencia de una gran variedad de moléculas orgánicas. Dado que una cantidad importante de polvo cometario se depositó con suavidad en la Tierra, su contenido orgánico pudo desempeñar un papel importante en los procesos prebióticos previos a la aparición de microorganismos. Esta revisión se centra en la descripción de los compuestos orgánicos de cometas y polvo cometario, así como su posible relación con el origen de la vida. [**Int Microbiol** 2005; 8(1):5-12]

Palabras clave: cometas · polvo cometario · polvo interplanetario · química prebiótica · evolución química · origen de la vida

Matéria orgânica em cometas e em pó cometário

Resumo. Os cometas são conglomerados primitivos do sistema solar que contêm uma mistura de gases gelados, partículas refratárias e partículas carbonáceas, ricas em elementos biogênicos. A espetacular aparência dos cometas se deve, principalmente a uma nuvem de partículas micrométricas de pó que abandona o núcleo do cometa a medida que os gases gelados se sublimam ao se aproximar do sol. Tanto a análise do pó do cometa capturado na estratosfera como as missões espaciais a cometas têm demonstrado a presença de uma grande variedade de moléculas orgânicas. Uma vez que uma quantidade importante do pó cometário se depositou com suavidade na Terra, seu conteúdo orgânico pode ter desempenhado um papel nos processos prebióticos, anterior ao aparecimento dos microrganismos. Esta revisão é centrada na descrição dos compostos orgânicos de cometas e do pó cometário, assim como sua possível relação com a origem da vida.[**Int Microbiol** 2005; 8(1):5-12]

Palavras-chave: cometas · pó cometário · pó interplanetário · química prebiótica · evolução química · origem da vida