COMETARY CHEMISTRY

Kathrin Altwegg

Celestial messengers carry a link between interstellar matter and life on Earth.

Ithough comets can appear extremely bright to observers, cometary nuclei are the darkest objects in our solar system. Their albedos are around just 4%. Their apparent brightness is caused by solar light scattering from their tails—micrometer-sized dust grains ejected from the nuclei. Although the nuclei are tiny, just a few kilometers in diameter, their visible tails can span the night sky, as seen here for comet McNaught behind Mount Paranal in Chile in 2007.

Despite their small size, comets receive significant research attention. Since ancient times, they have been recognized as special—and often as messengers announcing imminent doom. Today we know that comets actually carry a wealth of information about the solar system's early history.

To the best of our knowledge, comets formed far from the young Sun in a cold part of the protosolar disk. They were then scattered by the giant outer planets either into the Kuiper belt, a ring of objects that lies outside Pluto's orbit but still in the ecliptic plane, or into the isotropic Oort cloud, a collection of objects understood to reside 20000 AU from the Sun. And there they remain unless collisions or other gravitational disturbances scatter them to the inner solar system.

Temperatures in cometary reservoirs are below 30 K, so comets contain well-preserved material from the earliest stage of solar-system formation. When they warm upon entering the inner solar system, they shed their icy surface layers through sublimation, which continually exposes fresh material until the comets break up or lose all their volatile material and become dormant. That fresh mate-

rial dates back to the solar system's formation and can provide evidence of nucleosynthesis, the formation of complex organic molecules, and other chemical processes that happened along the way.

Cometary exploration

Remote sensing of cometary tails and comae, the atmospheres around comet nuclei, started with the appearance of Halley's comet in 1910 (see figure 1). The first *in situ* exploration, however, came when the European Space Agency (ESA) launched the *Giotto* spacecraft, which approached comet 1P/Halley during its 1986 return. The spacecraft's observations showed that comets are much more than dirty snowballs, as postulated by Fred Whipple¹ in 1950. Rather, they contain a plethora of molecules with atomic masses of 100 or more.

The appearances of two big comets—Hale–Bopp and Hyakutake in 1995 and 1996, respectively—led to a major step forward in cometary research. In the years since Halley's transit in 1986, Earth-bound telescopes have become more powerful, capable of covering wider frequency ranges and detecting many new chemical species in the bright



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comets' comae. Those telescopes now can also observe a larger population of comets—so many that ones from different families, such as long-period Oort-cloud comets (OCs) and short-period Jupiter-family comets (JFCs), can now be compared.²

Such species as diatomic nitrogen remain elusive, however, as they lack or have only weak optical transition lines. And even the most powerful telescopes observe mostly comets with strong outgassing—that is, active comets that travel relatively close to the Sun. Data therefore show a strong bias toward more active OCs relative to JFCs and toward comets with small perihelion distances.

In situ observations have come mostly from flyby missions targeting one or two comets at a predetermined distance from the Sun; for example, *Giotto* encountered comets 1P/Halley in 1986 and 26P/Grigg–Skjellerup in 1992. Missions are generally limited to flybys because of the high velocities the spacecraft would have to reach to keep up with the comets. Only the ESA's *Rosetta* probe³ followed a comet, 67P/Churyumov–Gerasimenko, over a large part of its orbit around the Sun (see figure 2). It launched in 2004 and, after three flybys of Earth and one of Mars, reached comet 67P/Churyumov–Gerasimenko in 2014. *Rosetta* stayed within a few kilometers of the comet for more than two years and finally crash landed on its surface in September 2016.

Rosetta carried 11 instruments and the landing unit *Philae*. It was the first probe to monitor a coma's evolution. Several instruments analyzed refractory material—the size, mass, speed, and composition of dust. Others were optical spectrometers that measured wavelengths from UV to microwave, and some, of course, were cameras. *Philae* itself had nine instruments on board, including two mass spectrometers and a drill.

Because of the comet's small size and low density, it exerts gravitational forces that are one hundred-thousandth the size of Earth's, so harpoons were used to anchor the lander to the surface. But they malfunctioned, and *Philae* hopped around, landing not once but four times. It ended up in a tilted position in the shadow of a ledge. Results from *Philae* are therefore scarce, but nonetheless extremely important, as they provide some ground truth. A summary of the mission and its main scientific results can be found in reference 4.

Why study comets?

Cometary material originated well before the solar system's formation 4.6 billion years ago. It contains tracers of all stages of formation, from the interstellar medium to planets to, maybe, the origin of life.

Figure 3 shows a sketch of the developmental stages of material in the solar system. The tenuous interstellar medium, with a density of less than 10³ cm⁻³, is composed of gas and dust released from long-dead stars, such as supernovae and red giants. Those objects are responsible for the nucleosynthesis of atoms heavier than helium and for producing submicrometer refractory grains composed of silicates, graphite, aluminum oxides, and other minerals. Each stellar source produces a specific isotopic signature.

Chemical reactions eventually arise in the interstellar medium, albeit slowly because of the low densities and temperatures. Simple gas-phase molecules, such as diatomic hydrogen, cyanide, and hydroxide, start to form. Dark molecular clouds—dense agglomerates of dust grains and gas—form by gravitational forces. They can be huge, up to 600 light-years in

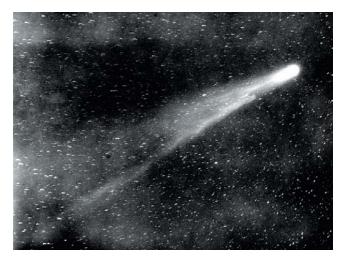


FIGURE 1. COMET 1P/HALLEY was first photographed during its 1910 traverse. (Courtesy of the Yerkes Observatory/public domain.)

diameter and 100 million solar masses. At $10^3\,\mathrm{cm}^{-3}$ and 10–20 K, the clouds are slightly denser and warmer than the surrounding interstellar medium.

Although interstellar-medium chemistry is governed by ion-neutral reactions in the gas phase, the ionization in dark clouds is low because of the absorption of radiation by dust. The tiny dust grains can act as catalysts for chemical reactions: Atoms or radicals condense onto the grains and then become joined. For example, carbon monoxide condenses and, over time, is converted to methanol with the addition of hydrogen. Dust grains therefore accumulate icy layers composed of different molecules. Condensation and subsequent sublimation and recondensation, which may be mass dependent and are driven by changes in surrounding radiation fields, cause isotopic fractionation.

Once gravitational forces collapse part of the dark cloud into a disk, the density increases as material flows toward the disk's center. Eventually it becomes high enough for a star to start forming. Temperatures in such regions show strong gradients. Before a star ignites, temperatures are around 10–20 K a few astronomical units from the protostellar disk's center along the midplane and 100 K in the densest part. Molecules near the center can sublimate from the icy layers of the dust grains, undergo chemical transformation, and then flow out into colder regions and recondense, leading to even more complex species.

Even after a protostar has formed and ignited, it's shielded by dust, so the midplane of the protoplanetary disk just a few astronomical units away remains dark and cold. Along the *z*-axis, however, bipolar jets originating from the star extend perpendicular to the plane and allow ions and hot material to mix with cold midplane material. That process may lead to additional chemical reactions, although the extent to which it does is still debated.

Material then aggregates into planetesimals from which planets form. Depending on a planetesimal's size and its distance from the star, any memory of the material's origin may already be partly or mostly lost. The material has already undergone sublimation, ionization, and chemical interactions with other species before recondensing. Large planetesimals are subject to considerable heating by radioactivity—mainly from

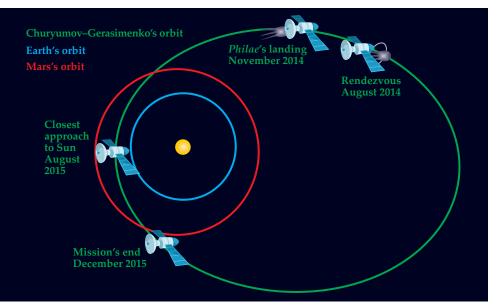


FIGURE 2. THE ROSETTA PROBE met comet 67P/Churyumov–Gerasimenko in 2014 when it was 3.8 AU from the Sun. It followed the comet for more than two years, including going through the comet's perihelion at 1.29 AU. (Adapted from a schematic by the ESA.)

the decay of ²⁶Al—which leads to liquid water in their interiors that can alter their minerals. The liquid phase also leads to differentiation, particularly in planets, big asteroids, and moons, with denser material sinking to the center. Small asteroids are often fragments of larger bodies and show signs of high-temperature processes with liquid water. The nature of the original material from which the solar system emerged is therefore nearly impossible to detect in planets, moons, and even asteroids.

In comets, however, evidence from the early solar system is preserved because the bodies experience little chemical activity in the solar nebula. Comets form in the outer protoplanetary disk, probably 30–50 AU from the center, and their sizes remain so small—a few kilometers in diameter—that heating by radioactivity is moderate at most. No aqueous alterations can be found in cometary material, although some high-temperature minerals have been detected, which is proof of some mixing with the protoplanetary disk. Comets' porous structures also have very low thermal inertia, which keeps their interiors cold.

Blast from the past

Once a comet enters the inner solar system, and during each orbit around the Sun, it sheds its damaged surface layer and exposes fresh, unaltered material. That material originates from the prestellar stages and can be sampled remotely or *in situ*. Comets also transport material from the cold outer edge of the solar system to the inner parts, including Earth. From the number of craters on our moon, it is evident that small bodies did impact planets and moons in quite large numbers well after the larger bodies formed and may therefore have changed or complemented terrestrial material by supplying water and organics.

Noble gases in cometary material undergo few chemical interactions, which makes the gases important for understanding how the material that makes up our solar system evolved. Their isotopic fingerprints reflect the earliest stage at which

the elements formed by nucleosynthesis and point to certain stellar origins. The abundances of volatile molecules, such as CO, methane, argon, and diatomic sulfur, present a record of the temperatures experienced by the material throughout its evolution.

Isotopologues—molecules with the same chemical formula that differ by at least one isotope, such as C¹6O, ¹³CO, and C¹8O—have different sublimation and condensation rates that lead to isotope fractionation. The rates and temperature dependences of their chemical reactions are also often slightly different, which again leads to fractionation. Studying isotopologues of different molecules provides clues about the molecules' formation temperatures. It also points to the kind of cold-temperature chemistry that may have occurred. Detecting evidence of such

processes as gas-phase, gas-grain surface, and ion chemistry provides information about the stage at which the compounds were formed.

The deuterium-to-hydrogen ratios in cometary water (HDO/H_2O) and $D_2O/HDO)$ are sensitive to external factors, including temperature and the type of chemistry that produced the water. Comparing those ratios in terrestrial and cometary water indicates how much water could have been delivered to Earth by comets.

Comparing the degree of organic-molecule complexity in comets with that in the interstellar medium, dark clouds, and star-forming regions hints at how much chemistry is inherited from those cold prestellar environments rather than from the protoplanetary disk. Prebiotic molecules and other species essential for life that are found in comets might provide clues about how life got started on Earth.

A cometary portrait

The comet 67P/Churyumov–Gerasimenko is a JFC with a period of 6.45 years, an aphelion distance of 5.68 AU, and a perihelion distance at 1.24 AU. It has a mass of about 10¹³ kg and a density of 533 kg m⁻³. The comet has a bilobed shape—its longest dimension is 4.3 km—that suggests it was once two cometesimals that gently collided. When *Rosetta* arrived in 2014, 67P was spinning with a period of 12.4 hours around an axis tilted relative to the ecliptic. Over the following two years, the comet's period decreased to 12.0 hours, a result of non-gravitational forces from its sublimating gas.

Rosetta's CONSERT and radio science instruments probed the interior of 67P's nucleus. Those data point to a homogeneous mixture of ice and dust with a density slightly higher at the surface than in the interior. It is unclear whether ice and dust are intimately mixed or the ice fills the voids between the dust grains. Water is scarce on the comet's surface—the temperature on the sunlit parts is around 200 K, even at large heliocentric distances. But the body's high porosity, about 75%, causes a huge thermal gradient. Subsurface temperatures can reach 120–160 K just a few millimeters to centimeters below the surface, where ice sublimation happens.

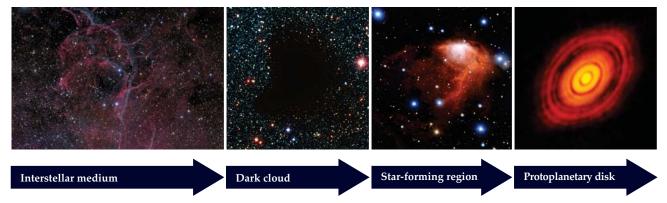


FIGURE 3. A STELLAR SYSTEM'S DEVELOPMENT starts with the interstellar medium. Once the material starts undergoing chemical reactions, it aggregates through gravitational forces and forms dark clouds, which eventually become dense enough to form stars surrounded by disks of protoplanetary material. That material comes together to form planets where life can potentially evolve. Information about all those stages can be gleaned from cometary material. Pictured from left are the Vela supernova remnant (courtesy of ESO/José Joaquín Pérez); the dark cloud B68 (courtesy of ESO); the star-forming cloud RCW 34 (courtesy of ESO); the protoplanetary disk around HL Tauri (courtesy of ALMA/ESO/NAOJ/NRAO); the solar system (image by Withan Tor/Shutterstock.com); and compounds that enable life (courtesy of ESA).

During 67P's perihelion passage, Rosetta witnessed many violent, short-lived—less than a half hour—outbursts of cometary material (see figure 4). The mechanism for those outbursts is not yet fully understood. Over the course of its orbit, the comet lost 0.1% of its mass by sublimation of ice and dust carried away by gas drag. Half of that measured mass loss was from volatiles, which means the comet lost an equivalent amount of dust. But ejected dust may not be lost; it can fall back onto the nucleus. That process was observed in many images of the comet's dust-covered northern hemisphere, where dust ejected from the southern hemisphere during perihelion returned to the surface. It's therefore hard to say whether comets are dirty snowballs, icy dirtballs, or, most likely, a mix of ice and refractory dust grains in approximately equal amounts.

Dust grains observed in the coma range from micrometers to decimeters in size.⁵ They are agglomerates made of subunits that can be as small as 100 nm. On impact, the agglomerates often fragment, a sign of their low tensile strength. They are composed of about equal parts minerals and organic refractory macromolecules. Those observations point to a gentle agglomeration of dust and ice at comet formation.

In situ mass spectrometry by Rosetta's ROSINA instrument yielded the most sensitive analysis to date of the chemical composition of a comet's icy part. In particular, the high mass resolution (9000 at m/z = 28) and sensitivity of the double-focusing mass spectrometer, a classical magnetic spectrometer in Nier-Johnson configuration, led to many detections of molecules and isotopologues never seen before in cometary comae. 6 More than

66 parent molecules, including some minor isotopologues, could be identified. Data analysis is still ongoing, and more results from the mission are expected.

Chemical composition

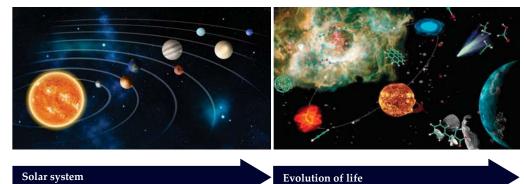
One of the biggest surprises from Rosetta was the detection of abundant O₂, which is highly volatile, is extremely chemically active, and should not survive as a gas in such hydrogendominated environments as the solar nebula and protoplanetary disks. Strikingly, in 67P, the O₂ sublimation followed the same trajectory as H₂O sublimation, which occurs around 140 K. Other highly volatile chemicals instead followed that of CO₂ and had much lower sublimation temperatures, around 60 K. The different sublimation temperatures were evident from large compositional inhomogeneities in the coma, where the CO₂/H₂O ratio changed by a factor of 20 but the O₂/H₂O ratio remained constant.

Several mechanisms, such as the radiolysis of water ice, were proposed to explain the formation of O₂ and the constant ratio of the highly volatile O₂ to H₂O. But the ¹⁶O/¹⁸O ratios of H₂O and O₂ are incompatible,⁷ which leaves only a primordial origin for the O_2 , either in the gas phase or on the surfaces of grains. The O₂ was then incorporated in a water-ice matrix of dust grains and never sublimated before it ended up in the coma of 67P.

Rosetta also detected the first neutral N2, another molecule that is hard to detect remotely because it lacks a dipole moment. But the amount of N₂ found does not explain the missing nitrogen that had already been noticed during the Giotto mis-



from comet 67P/Churyumov-Gerasimenko was captured by Rosetta's OSIRIS narrowangle camera on 29 July 2015. No signs of the jet are seen in the left image, taken at 13:06 Greenwich Mean Time (GMT). It is strong in the middle image, captured at 13:24 GMT. Residual traces are only faintly visible in the right image, taken at 13:42 GMT. (Courtesy of ESA and the OSIRIS



sion. Comets seem to have less nitrogen than would be expected based on its cosmic abundance. Nevertheless, the detection of N₂, together with CO, CH₄, and Ar, shows that comets accumulate frozen, highly volatile species from their environments. That means temperatures during comet formation would have been around 25 K.

Molecular formation

Isotopes have different fingerprints depending on what stellar environment-for example, a low-mass star, supernova, or neutron-star merger - hosted their nucleosynthesis. Those fingerprints can change or even disappear, however, because sublimation, condensation, and other chemical reactions often have small but nonnegligible isotopic dependences, especially at the cold temperatures encountered in space. Still, comparing the isotopic ratios of different chemical species can provide clues about the physical and chemical conditions in which the species formed.

Noble gases undergo few chemical reactions. They there-

high amount of 129Xe can be explained as the decay product of iodine-129, which has a lifetime of 1.57 x 107 years. To get that result, the solar nebula was probably poorly mixed when 67P and other comets formed; if it was, the isotopic makeup of cometary Xe should be the same as solar Xe. The Xe isotopic ratios on Earth highlight a long-standing

puzzle about the element's sources. Earth has two Xe reservoirs: its interior and its atmosphere. The material inside Earth is chondritic, meaning its makeup is consistent with that of stony, nonmetallic chondrites-meteorites that have not been modified by melting or differentiation. The atmospheric reservoir, however, is neither chondritic nor solar. Over time, Earth has lost Xe, mostly light isotopes, from its atmosphere to space. But even after correcting for that fractionation, the atmosphere is unexpectedly depleted of heavy Xe isotopes.

Mixing $(22 \pm 5)\%$ cometary Xe with chondritic Xe yields the isotopic ratios of Earth's atmospheric Xe. Achieving that composition would require roughly 100000 cometary impacts. With that number of impacts and considering the ratio of Xe to water in comets, researchers have concluded that less than 1%

> of today's surface water was brought by comets. The quantity of organics relative to Xe in 67P, however, indicates that the amount of organic matter brought to Earth by comets surpasses today's biomass. That enormous amount of organics may have helped to spark life on Earth.

fore retain the best-preserved isotopic distribution from nucleosynthesis. A good example is xenon, which has nine stable isotopes. One of the most important results from the ROSINA instrument is the isotopic distribution⁸ of Xe in 67P. Compared with the solar distribution, it is depleted in the heavy isotopes ¹³⁴Xe and ¹³⁶Xe

(see figure 5). The relatively

Other evidence supporting the idea that most of Earth's water did not originate in comets comes from the D/H ratio, which is mostly a product of chemical fractionation. Earth's water is enriched by a factor of 10 compared with the interstellar medium and the Sun, which have D/H ratios of 1.5×10^{-5} . Still, researchers already realized from the Giotto mission to Halley that comets have a higher D/H ratio than Earth, and 67P has a value of 5.3×10^{-4} , even higher than the 3×10^{-4} of Halley and other Oort-cloud comets. The average value for all comets measured so far is 3.6×10^{-4} , which makes it highly unlikely that most terrestrial water comes from comets, as was once postulated.

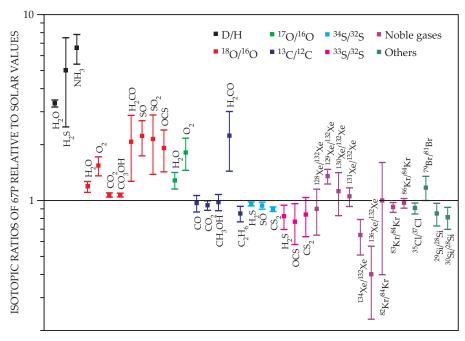


FIGURE 5. ISOTOPIC RATIOS of elements and compounds found in comet 67P/Churyumov-Gerasimenko differ from solar values. The deuterium/hydrogen ratios are shown relative to terrestrial values. (Courtesy of Rosetta's ROSINA team.)



Doubly deuterated water, D_2O , is abundant in 67P compared with what would be statistically expected. The fraction of deuterated water that is doubly deuterated is compatible with what is measured in the interstellar medium. But the fraction of water that is deuterated is nearly 70 times as high as would be expected at thermal equilibrium. That means the comet's water is the result of highly nonequilibrated chemistry that could only have happened in the extremely cold interstellar medium. The comet's ice was therefore inherited as solid ice from the prestellar stage and never released into the gas phase, a process that would have immediately lowered the ratio by isotopic exchange reactions with the gas-phase hydrogen, which has a low D/H ratio.

As seen in figure 5, the isotopic ratios of many elements in 67P deviate from solar values by at least one standard deviation. Silicon's heavier isotopes are underrepresented, another hint that the solar nebula was not well mixed. Sulfur is likewise depleted in heavy isotopes in all molecules that could be measured.

Oxygen isotopic ratios on the comet vary. Whereas CO, CO₂, and methanol show solar-like values, formaldehyde is enriched in ¹⁸O. Methanol's formation can be explained by grain-surface reactions, with CO freezing out of the gas phase onto grains and then being hydrogenated. The freezing enriches ¹⁸O in CO while leaving behind an ¹⁸O-depleted gas phase. But H₂CO is enriched in heavy oxygen and has a high ¹³C/¹²C ratio compared with methanol and CO, which means it cannot stem from an intermediate step in the CO hydrogenation process. Nor does a gas-phase-chemistry origin fit, as the H₂CO would then be depleted in the heavy isotopes. The riddle remains unsolved.

Water on 67P is slightly enriched in heavy oxygen (17 O and 18 O) compared with solar abundances, and the O_2 is more enriched in the heavy isotopes than the water. Self-shielding models 10 predict primordial water to be enriched in 18 O because of

FIGURE 6. THE COMETARY ZOO is filled with gases detected by the ROSINA instrument on *Rosetta*. Identification of complex molecules on 67P/Churyumov–Gerasimenko dispelled the myth that comets contain only simple substances and cast doubt on whether certain biosignatures could serve as evidence of life. For more details about the menagerie, see https://blogs.esa.int/rosetta/2016/09/29/the-cometary-zoo. (Adapted from an illustration by ESA.)

UV photodissociation of CO, whereas the solar wind is expected to be depleted in 18 O. The data therefore favor an interstellar origin for O_2 in comets and are not compatible with radiolysis of water ice, a commonly accepted source for O_2 on other bodies, such as Jupiter's moon Europa.

From stars to life on Earth

Although the mass spectrometers on *Giotto* were built to detect water and simple molecules with a limited mass range (1–56 Da) and resolution (m/ δ m \approx 40), the Picca instrument — an energy analyzer—detected masses up to 100 Da. The molecules' thermal velocities were well below the 68 km/s flyby speed, so species could be separated by their mass-dependent energies. The resolution was insufficient to identify the molecules, but it became clear that complex organic molecules were present in Halley's coma.

Rosetta's close proximity to 67P allowed it to detect many new species, shown in a cometary zoo in figure 6. Among them are several molecules that could participate in prebiotic chemistry. The detection of long carbon chains of up to seven carbons (giraffes), aromatic hydrocarbons (elephants), oxygenated hydrocarbons (exotic birds and monkeys), and a diverse population of sulfur-bearing molecules (skunks and frogs) changed the perception that comets contain only simple molecules like CO, CO₂, NH₃, and water.

One of Rosetta's highlights was the lion-glycine, the simplest amino acid. Such complexity probably stems mostly from the presolar stages of the solar system, specifically from dark clouds and star-forming regions. The abundances of cometary parent molecules and those observed in the interstellar medium show a striking similarity for many of the species.⁷

Recently, traces of ammonium salts of the form NH₄R⁻(saltwater crocodiles) have been detected in ROSINA data.¹² The salts are formed in reactions between ammonia and acids such as hydrogen cyanide, hydrochloric acid, and formic acid. Their sublimation temperatures are higher than those of the individual parts, and upon sublimation, they mostly dissociate again into NH₃ and acid. The presence of ammonium salts provides a likely explanation for the missing nitrogen, as they lock nitrogen in a refractory state in which it escapes detection.

Ammonium salts also have astrobiological relevance: They are involved in the formation of amino acids and of the nucleobase adenine from NH₄CN. The salts form cyanamide, which then reacts with glycolaldehyde to form natural nucleotides. Both prebiotic molecules—cyanamide and glycolaldehyde were found in the coma of 67P.

Another important result is the detection of phosphorus monoxide. Early Earth had plenty of phosphorus, but most was probably tied up in minerals and therefore not available for biological processes. But life as we know it needs phosphorus for, among other things, DNA and the energy carrier ATP (adenosine triphosphate). Recently, an international collaboration of researchers sketched the cosmic journey of soluble phosphorus from a massive supernova where phosphorus is formed by nucleosynthesis, to the observation of PO in starforming regions, to comets where PO is enclosed in the cometary ice, and -potentially - to Earth, where it was a necessary component for life to start.¹³

The material in comets is not specific to the solar system or Earth. The processes that happened here can happen everywhere. Thousands of planets have been found orbiting a wide variety of stars, and those discoveries have triggered significant interest in finding life elsewhere in the universe. The Rosetta mission's results make that search more difficult: Molecules that could serve as biosignatures, such as O2 together with CH4 or amino acids, are now known to exist in the nonliving world of cometary ice. Such biosignatures are therefore insufficient evidence for life on a planet, since they can have a nonbiogenic origin. Still, knowing that complex prebiotic molecules exist and how they could be delivered to planets may help focus the search for life.

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