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COSMOCHEMISTRY

Xenon isotopes in 67P/Churyumov-Gerasimenko show that comets contributed to Earth's atmosphere

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The origin of cometary matter and the potential contribution of comets to inner-planet atmospheres are long-standing problems. During a series of dedicated low-altitude orbits, the Rosetta Orbiter Spectrometer for Ion and Neutral Analysis (ROSINA) on the Rosetta spacecraft analyzed the isotopes of xenon in the coma of comet 67P/Churyumov-Gerasimenko. The xenon isotopic composition shows deficits in heavy xenon isotopes and matches that of a primordial atmospheric component. The present-day Earth atmosphere contains $22 \pm 5\%$ cometary xenon, in addition to chondritic (or solar) xenon.

Comets are among the most pristine solar system materials (1). Their abundant volatile species, mainly in the form of ices, are intimately mixed with refractory silicate-rich phases and organics, the origins of which—either in the protosolar disk or molecular clouds and interstellar medium—are still under debate (1). The discovery that Comet 81P/Wild contains high-temperature minerals akin to those found in primitive meteorites with key isotopic signatures (e.g., that of oxygen) typical of solar system reservoirs (2) suggests that some of the cometary constituents were cycled close to the proto-Sun and were radially transported outward. However, the case of ice is less clear. Enrichments in deuterium and nitrogen-15 in comae (3, 4) have been regarded as either originating from processing in the disk outskirts under irradiation or resulting from low-temperature ion-molecule reactions in molecular clouds. Another unresolved problem is the possible contribution of comets to inner-planet atmospheres. Although the D/H and $^{15}\text{N}/^{14}\text{N}$ signatures of the terrestrial atmosphere and oceans suggest an inner solar system origin for volatile elements on Earth (5), variations in the D/H ratio in primitive meteorites point to the contribution of interstellar water to asteroids (6).

Xenon, the heaviest stable noble gas, with nine isotopes of different nucleosynthetic origins (7, 8), is a key element for identifying nuclear components present in presolar material. The composition of solar system Xe, represented by

measurements of the solar wind (7), is the result of the homogenization of such components. Several radioactive decays also produce Xe isotopes, among which that of ^{129}I (half-life of 15.6 million years) decaying into ^{129}Xe provides time constraints on the early evolution of the solar system. Earth's atmosphere has a xenon composition that is unique among solar system objects and reservoirs (9). It is depleted relative to expectations based on the extrapolated behavior of the lighter noble gases Ne, Ar, and Kr; by a factor of ~ 20 relative to Kr (normalized to the abundance in chondritic meteorites). Atmospheric Xe is also isotopically (mass-dependently) fractionated, being enriched in heavy isotopes by 30 to 40 per mil per atomic mass unit (u) compared with chondritic xenon (hereafter, Q-Xe) or solar wind xenon (hereafter, SW-Xe); this is known as the xenon paradox (10). When corrected for mass-dependent isotopic fractionation (MDF), atmospheric Xe does not directly correspond to any known solar system component. These observations have led to the definition of a theoretical primordial component termed U-Xe (9) (where U stands for Ur; the German word for primordial, and not for uranium-derived fission Xe). U-Xe is close to SW-Xe for the $^{124}\text{--}^{130}\text{Xe}$ isotopes but is depleted in the heavy Xe isotopes, particularly ^{134}Xe and ^{136}Xe . U-Xe has not been clearly identified in meteorites or planetary samples.

We report the determination of the isotopic composition of xenon in a comet. Xenon isotopes

were measured with the Double Focusing Mass Spectrometer (DFMS) of the Rosetta Orbiter Spectrometer for Ion and Neutral Analysis (ROSINA) instrument suite (11) on the Rosetta spacecraft. DFMS is a high-mass-resolution instrument (mass divided by change in mass = 3000 at 1% peak height at a mass/charge ratio of 28 u/e) that measured gases emitted by comet 67P/Churyumov-Gerasimenko (67P/C-G), presumably from sublimation of ice (3). The Xe measurements were carried out during a series of dedicated low-altitude orbits, between 10 and 7 km from the comet's center of mass, from 14 to 31 May 2016 (12). Because of the high resolution of DFMS, mass/charge ratios of 129, 131, 132, 134, and 136 u/e were essentially free of interfering species of similar mass, as confirmed by the shapes of the peaks, whereas interference presumably due to S_4^+ was detected at mass/charge = 128 u/e and corrected using peak deconvolution (12). A slight contribution of a few percent at mass/charge = 130 u/e, due to S_4^+ -containing ^{34}S , was also corrected. The average Xe isotope ratios were derived from this database, with uncertainties corresponding to 1 standard deviation (σ) of the mean. The ROSINA instrument was equipped with a gas calibration unit that permitted in-flight analysis of reference gases, including xenon (12).

Figure 1 shows the xenon composition of 67P/C-G, normalized to that of the solar wind (horizontal orange line) and to ^{132}Xe (the most abundant Xe isotope in most cases), together with other solar system compositions (terrestrial atmosphere and chondritic). The data obtained from the in-flight calibration runs are consistent with a terrestrial Xe composition, as expected, but the 67P/C-G Xe isotopic ratios deviate markedly from solar, or chondritic, values. Whereas $^{128}\text{Xe}/^{132}\text{Xe}$, $^{130}\text{Xe}/^{132}\text{Xe}$, and $^{131}\text{Xe}/^{132}\text{Xe}$ are solar-like within uncertainties, 67P/C-G Xe is strongly depleted in ^{134}Xe and ^{136}Xe , by ~ 40 and $\sim 60\%$, respectively.

We first tested the possibility that the observed variations are due to MDF, in which case the data should align along the dotted curve of Fig. 1. The $^{129\text{--}136}\text{Xe}/^{132}\text{Xe}$ ratios could be reasonably accounted for by MDF affecting a solar-like Xe component. The required fractionation factor, however, should be extremely high, around 14% per atomic mass unit, higher than ever observed for Xe (or other noble gases) in any solar system object or reservoir. Notesco *et al.* (13) found no evidence for Xe isotopic fractionation upon trapping in water ice, and trapping of ionized Xe results in a MDF of no more than 1% (14). Alternatively, MDF could also be due to distillation enhancing isotopic fractionation during loss of Xe isotopes from a reservoir (e.g., cometary ice). To produce

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Fig. 1. Xenon isotopic composition of comet 67P/C-G's volatile fraction.

Xe isotopes (^{i}Xe) were analyzed by the ROSINA DFMS on the Rosetta spacecraft. They are normalized to ^{132}Xe (ratios shown by the blue circles and line) and the solar wind composition (7) (horizontal orange line). Error bars, 1σ . The chondritic (26) (Q-Xe; green line) and atmospheric (10) (light blue line) compositions are also shown for comparison. The light blue diamonds show data obtained from in-flight calibration using a terrestrial Xe reference gas (12). The black dotted curve represents the effect of mass-dependent isotopic fractionation (MDF), for which a fractionation factor of 14% per atomic mass unit was adjusted to fit the observed $^{129-136}\text{Xe}/^{132}\text{Xe}$ isotope ratios. Three MDF processes (kinetic, equilibrium, and gravity and centrifugal force) were considered and yielded curves that are indistinguishable from each other at the scale of the figure (12).

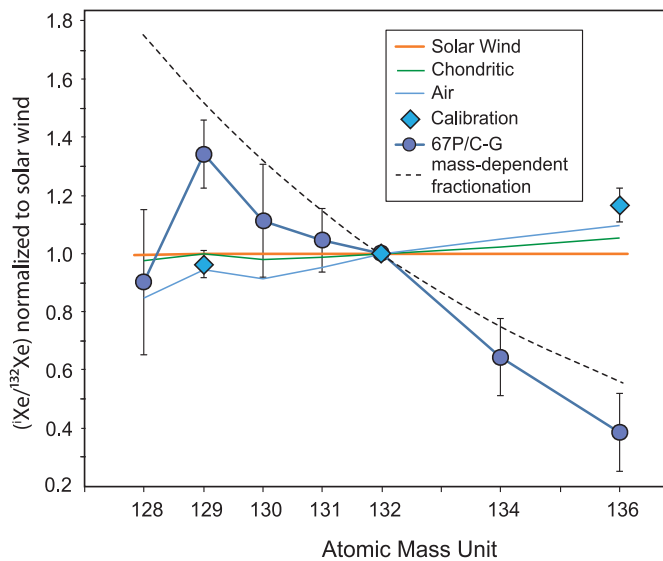
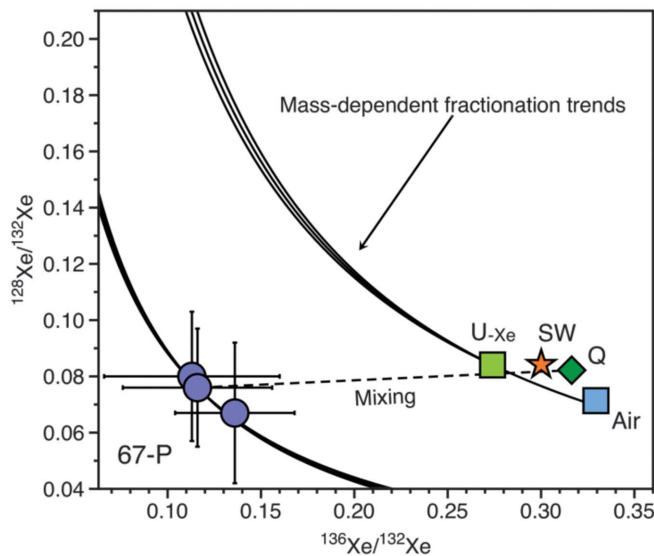


Fig. 2. Testing the origins of Xe isotope variations in comet 67P/C-G.

The Xe isotopes are normalized to ^{132}Xe and the solar wind composition (7). Error bars, 1σ . We tested the possibility that 67P/C-G Xe is derived from MDF of primordial (U), solar wind (SW), or chondritic (Q) Xe. Three MDF processes are envisioned (12), yielding slightly different MDF curves (solid curves). None of these can reproduce 67P/C-G Xe from U-Xe, SW-Xe, or Q-Xe compositions.

Likewise, none of the MDF curves from 67P/C-G Xe can reproduce U-Xe, SW-Xe, or Q-Xe compositions. Mixing of Q-Xe or SW-Xe with 67P/C-G Xe is able to reproduce the composition of U-Xe (Fig. 4).



a MDF of 10% per atomic mass unit would require Xe loss on the order of 10^4 for an instantaneous fractionation factor of 1%. Such Xe depletion is not consistent with the detection of xenon in the coma of 67P/C-G, nor with the $^{132}\text{Xe}/^{36}\text{Ar}$ ratio of 2.5×10^{-2} estimated for the comet from our DFMS measurements [much higher than the solar ratio of 1.8×10^{-5} (15)]. Furthermore, degassing of a comet is essentially a one-way process in which a portion of fresh ice exposed to solar heating is sublimated, removing water vapor and volatiles trapped in ice to space. Last, xenon iso-

topes were analyzed over several weeks from several degassing sources without showing substantial variation (12), and it is unlikely that the recorded composition could correspond to a fractionated subreservoir. Thus, the Xe isotopic fractionation should have occurred before trapping in cometary ice, a possibility that cannot be discarded, given our poor knowledge of processes having taken place in the outer solar system.

The $^{128}\text{Xe}/^{132}\text{Xe}$ ratio does not fit into the isotopic fractionation scenario (Figs. 1 and 2). In Fig. 2, we show the results of testing whether

the composition of 67P/C-G Xe can be derived from MDF of solar or meteoritic Xe. Three MDF processes are envisioned (kinetic, equilibrium, and gravity and centrifugal force) (12), yielding slightly different MDF trends, and none of them can reproduce 67P/C-G Xe from U-Xe, SW-Xe, or Q-Xe compositions. In contrast, mixing of Q-Xe or SW-Xe with 67P/C-G Xe reproduces the composition of U-Xe (Fig. 2).

Solar system xenon is the result of mixing of three different nucleosynthetic sources: the p-process, producing the rare ^{124}Xe and ^{126}Xe isotopes (which could not be measured here); the s-process, producing $^{128-132}\text{Xe}$ isotopes; and the r-process, producing $^{134,136}\text{Xe}$ isotopes (8). Thus, the isotopic composition of cometary xenon is possibly the result of a nucleosynthetic mix that is different from that which produced solar xenon. From theoretical considerations and correlations of Xe isotopes observed in presolar grains [SiC and nanodiamonds (8)], the s-process Xe composition is reasonably well defined, but Xe isotope variations in presolar material require at least two different r-process compositions (8). Figure 3 shows mixing of 35% s-process Xe with 65% r-process Xe (12), the latter being defined by the two r-process compositions identified by (8).

This scenario accounts reasonably well for the deficits of ^{134}Xe and ^{136}Xe observed in 67P/C-G and is more consistent with the cometary $^{128}\text{Xe}/^{132}\text{Xe}$ ratio than the MDF model is. It implies that ^{129}Xe is enriched relative to ^{132}Xe and the solar composition by $34 \pm 11\%$ (1σ). This large monoisotopic excess could be due to a specific nucleosynthetic process preferentially producing ^{129}Xe , a possibility supported by some nanodiamond fractions with $^{129}\text{Xe}/^{132}\text{Xe}$ ratios up to 2.2 (16). Alternatively, in situ (within ice) β -decay of ^{129}I (half-life of 15.6 million years) after the start of solar system formation (SSSF) is unlikely: The $^{129}\text{I}/^{127}\text{I}$ ratio at the SSSF was $\sim 1 \times 10^{-4}$ (17), and an in situ origin after the SSSF would require an enrichment of iodine relative to xenon by three to four orders of magnitude, which is not observed in the coma of 67P/C-G (12). This ^{129}Xe excess could then result from in situ (within ice) or ex situ (in the ambient gas) decay of ^{129}I before the SSSF. In this case, xenon trapped in ice would be much older, by several tens of millions of years, than the SSSF. Assuming that ^{129}I and ^{127}I were produced in comparable proportions during r-process nucleosynthesis, icy grains should have trapped both I and Xe relatively shortly after their last nucleosynthetic event. This exotic Xe component would have to be older than 100 million years before the SSSF, given the half-life of ^{129}I , which is consistent with a galactic evolution origin as constrained by the initial solar system abundances of ^{129}I , ^{244}Pu , and ^{237}Cm (18).

Cometary noble gases are concentrated in ice (13, 19, 20), and a presolar origin for Xe would imply that cometary ice is also presolar. Such an origin is in agreement with the detection of abundant O_2 (1 to 10% relative to H_2O) released by 67P/C-G (21). Indeed, molecular oxygen might

Fig. 3. Mixing of various nucleosynthetic components for the origin of 67P/C-G Xe.

The Xe isotopes are normalized to ^{132}Xe and the solar wind composition (7) (horizontal orange line). Error bars, 1σ . The red dots represent the best fit of mixing between s-process Xe and the two r-process end-member compositions identified by (8) (s-r1-r2; symbols are slightly displaced to the right for better visibility) [see (12) for methods]. The light brown area represents the field of possibilities for random contributions of the s-process and the two r-processes.

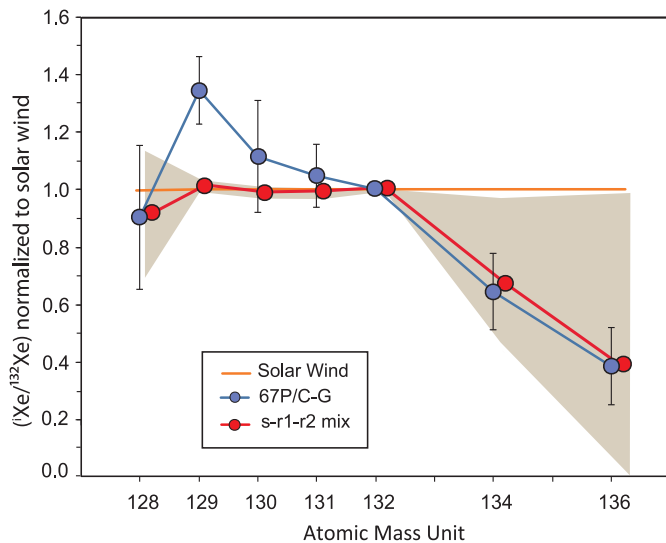
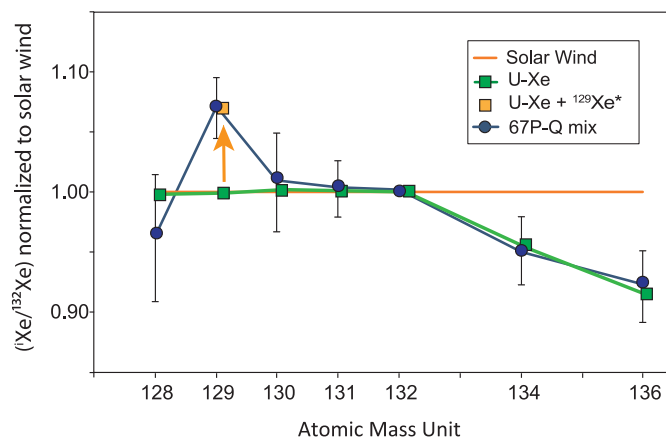


Fig. 4. A mix of chondritic and cometary Xe as the origin of a primordial atmospheric Xe component.

The Xe isotopes are normalized to ^{132}Xe and the solar wind composition (7) (horizontal orange line). Error bars, 1σ . Symbols for U-Xe (9) are slightly displaced to the right for better visibility. The U-Xe composition (green squares) is well reproduced by mixing cometary (67P/C-G) Xe with Q-Xe (67P-Q; dark blue dots). The cometary contribution, $22 \pm 5\%$, and the propagation of errors were determined using a Monte Carlo approach (12). The mixture is also able to reproduce the monoisotopic excess of ^{129}Xe observed in air (6.8%) (9), represented by the orange square.



have been produced from the radiolysis of H_2O icy grains by cosmic rays and/or hard photons before the comet was assembled (21), and this process would require long periods of time (10^7 to 10^9 years) in the protosolar nebula and/or low-density environments such as molecular clouds (22). A presolar origin is independently supported by the presence of S_2 in 67P/C-G (23) and $\text{HDO}/\text{H}_2\text{O}$ and $\text{D}_2\text{O}/\text{HDO}$ ratios closely resembling those of interstellar material (3).

The peculiar Xe isotopic composition of 67P/C-G provides a fingerprint of the contribution of comets to the terrestrial atmosphere. 67P/C-G Xe, which is largely depleted in ^{134}Xe and ^{136}Xe , indicates that comets may be the source of U-Xe. Mixing $22 \pm 5\%$ (1σ) cometary (67P/C-G) Xe with Q-Xe reproduces the composition of U-Xe

(Fig. 4). The choice of Q-Xe for the second component—instead of, for example, solar—is consistent with observations that (i) the H, N, and Ar isotopic compositions of the atmosphere and oceans are within the range of variations observed in chondrites and different from solar values (5, 7) and (ii) xenon in Earth's interior contains a chondritic component (24). A cometary contribution to atmospheric noble gases, suggested independently by modeling to be 19 to 27% (25), might have taken place during the proto-Earth growing stages (25) or later during the Hadean era (4.5 to 3.8 billion years ago) (20). Such a contribution would, however, have been minimal for the terrestrial oceans [$\leq 1\%$ (12)] and, by consequence, would not have affected the terrestrial D/H ratio.

A $22 \pm 5\%$ contribution of comets to atmospheric Xe also reproduces the $6.8 \pm 0.3\%$ (1σ) ^{129}Xe excess observed in the atmosphere (Fig. 4). This excess, classically attributed to the decay of radioactive ^{129}I trapped in the growing Earth, has previously allowed researchers to set time constraints on the accretion of Earth and the development of its atmosphere (17). Degassing of mantle Xe containing radiogenic ^{129}Xe through geological periods of time also contributed ^{129}Xe to the atmosphere, but probably no more than $\sim 1\%$ (12). Thus, a large fraction of the monoisotopic ^{129}Xe excess in the terrestrial atmosphere may be inherited, implying that the ^{129}I - ^{129}Xe system as a geochronological tool should be reconsidered and that the Wetherill's retention time of ~ 100 million years after the SSSF (17) should be seen as a lower limit. It remains unclear whether inherited cometary ^{129}Xe is consistent with the presence in the present-day Earth atmosphere of a component derived from the fission of ^{244}Pu .

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SUPPLEMENTARY MATERIALS

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Materials and Methods
Supplementary Text
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Xenon isotopes in 67P/Churyumov-Gerasimenko show that comets contributed to Earth's atmosphere

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Comets contributed to Earth's atmosphere

Models of xenon's origin in Earth's atmosphere require an additional, unknown source that has been a mystery for several decades. Marty *et al.* measured isotopic ratios of xenon released from comet 67P/Churyumov-Gerasimenko and found that they match the heretofore unknown source. The xenon appears to have been trapped in ice within the comet since before the solar system formed. Comets contributed about a quarter of the xenon on Earth, which constrains the amount of other materials (such as water) delivered to our planet by comets.

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