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Helium and Neon Abundances and Compositions in Cometary Matter

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Materials trapped and preserved in comets date from the earliest history of the solar system. Particles captured by the Stardust spacecraft from comet 81P/Wild 2 are indisputable cometary matter available for laboratory study. Here we report measurements of noble gases in Stardust material. Neon isotope ratios are within the range observed in “phase Q,” a ubiquitous, primitive organic carrier of noble gases in meteorites. Helium displays ³He/⁴He ratios twice those in phase Q and in Jupiter’s atmosphere. Abundances per gram are surprisingly large, suggesting implantation by ion irradiation. The gases are probably carried in high-temperature igneous grains similar to particles found in other Stardust studies. Collectively, the evidence points to gas acquisition in a hot, high ion-flux nebular environment close to the young Sun.

Comets are frozen, largely unaltered reservoirs of dust and gases present in the early solar nebula. They are likely to contain well-preserved records of the chemical, mineralogic, and isotopic character of primordial solar-system matter. On 15 January 2006, the Stardust Mission returned to Earth with a cargo of particles collected from the coma of comet 81P/Wild 2 (*1*). These not only open the door to detailed laboratory investigation of the nature of cometary matter, but will also provide chemical, mineralogic, and isotopic markers for identifying samples of comets suspected to be already present in extraterrestrial material collections [e.g., (*2–6*)].

Noble gases are excellent tracers of contributions from various solar-system volatile reservoirs and of physical processing of gases

acquired from these reservoirs. Their elemental and isotopic compositions in primitive meteorites differ from those in the Sun, as represented by the solar wind. Planetary atmospheres display noble gas signatures distinct from both solar and meteoritic patterns. A detailed knowledge of cometary noble gas abundances and isotopic compositions, unknown before Stardust, will allow investigation of compositional links between comets, the solar nebula, primitive meteorites for which a cometary origin has been advocated (*7, 8*), micrometeorites and interplanetary dust particles (IDPs), and the atmospheres of Earth, Mars, and Venus, where contributions of noble gases carried by comets have been proposed and debated (*9–11*).

The Wild 2 particles were collected by Stardust in aerogel, a porous low-density silica glass designed to decelerate an impacting grain with minimal alteration (*1*). The material analyzed in this study was extracted from the bulbous cavity wall of the capture track shown in Fig. 1A. We report results of helium and neon measurements on five subsamples of this material [Fig. 1B and fig. S1 (*12*)], carried out independently, using different extraction and analytical methods, at Centre de Recherches Pétrographiques et Géochimiques (CRPG) Nancy, France, and the University of Minnesota, Minneapolis, USA

(*13*). Gases were thermally released from the five samples, two (Thera-1 and Thera-2) at CRPG by single-step laser melting and three (St-1, St-2, St-3) at Minnesota by multistep pyrolysis to ~1400°C. Evolved He and Ne abundances were not large, in some cases exceeding blank levels by factors of only 2 to 3, and so accurate assessments of aerogel and instrumental blanks were crucial. Details on blank determinations, and other discussions of materials and methods, are set out in (*12*).

The bulbous track-wall samples are mixtures of aerogel and small grains shed from the impacting particle. Gases were held very retentively in their host grains; temperatures above ~1250°C (±50°C) were required to degas He and Ne in the Minnesota step-heating procedure. Originally we thought that gases might have been liberated by flash heating of the incident particle in the upper part of its deceleration track (Fig. 1A) and trapped in melted aerogel that rapidly chilled to silica glass along the track walls. It is now clear that the ~1200° to 1250°C temperatures reached before initiation of release (*12*) are incompatible with siting in aerogel. Diffusion coefficients for He in vitreous silica (*14*) are so large at these temperatures that glassy aerogel would empty itself in ~10 s—comparable to step heating times—even if diffusion distances were comparable to the 1-mm sample dimensions (Fig. 1B and fig. S1), and much more quickly (<1 s) at the ≤100-μm scale. Observed release profiles instead point to gas siting in refractory grains. This inference is consistent with the bulb-wall mineralogy measured by x-ray absorption spectroscopy (supporting online text), indicating that the grains are composed primarily (~75% by mass) of high-temperature metal (FeNi), metal-sulfur, and metal-carbon minerals. The remaining 25% are mostly silicates together with unidentified components. A separate search for carbon compounds was carried out by infrared (IR) spectroscopy (supporting online text) to address the possibility that the gas carriers might be refractory organic materials. None were detected. However, the search covered only a small fraction of the bulb wall and adjacent aerogel, so the presence of heterogeneously distributed organics is not ruled out. We return to the issue of the nature of the gas carrier later in the paper.

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Neon isotopic compositions were measured in both noble gas laboratories. Extractions with enough Ne for relatively precise measure-

ment yielded similar isotope ratios: $^{20}\text{Ne}/^{22}\text{Ne} = 10.68 \pm 0.35$ and 10.49 ± 0.24 together with $^{21}\text{Ne}/^{22}\text{Ne} = 0.0273 \pm 0.0024$ and $0.0279 \pm$

0.0017 , measured, respectively, in St-1 at Minnesota and Thera-2 at CRPG (Table 1). Both $^{20}\text{Ne}/^{22}\text{Ne}$ ratios are $>2.5\sigma$ above the atmospheric value of 9.80. These compositions fall within the range observed for meteoritic Ne—specifically, for Ne in “phase Q” (15), a minor macromolecular organic phase, ubiquitous in chondritic and achondritic meteorites, that hosts neon with $^{20}\text{Ne}/^{22}\text{Ne}$ ratios ranging from 10.1 to 10.7, and $^{21}\text{Ne}/^{22}\text{Ne}$ from a lower limit of 0.0278 to 0.0294 (Table 1). Stardust Ne lies within error in the Ne-Q data field, with the nominal $^{21}\text{Ne}/^{22}\text{Ne}$ ratios straddling its lower limit (Fig. 2A).

Are there alternatives to a Q-Ne-like source—in particular, a source with solar composition (16–18)—that might account for the Ne isotopic distributions within the uncertainties of the present data? The $^{20}\text{Ne}/^{22}\text{Ne}$ ratio in Thera-1 could be consistent with a solar-wind Ne end-member (Table 1), but it is not diagnostic because its large uncertainty also overlaps the air and Q compositions. Compared to solar wind ratios, $^{21}\text{Ne}/^{22}\text{Ne}$ in Thera-1 is lower by $\sim 3\sigma$, and the two most precise determinations of $^{20}\text{Ne}/^{22}\text{Ne}$ are $\sim 10\sigma$ lower (Table 1). Contemporary solar wind as a principal source of Stardust Ne is therefore ruled out, a conclusion reinforced by measured $^3\text{He}/^4\text{He}$ ratios that differ from those of the solar wind by $>5\sigma$ (Fig. 2B). One possibility might be that the Ne composition reflects mixing of solar Ne with an air contaminant. However, the source of such a large and similar air contribution in both laboratories—constituting $\sim 75\%$ of the ^{20}Ne in St-1 and Thera-2 and greatly exceeding aerogel blank levels (12)—is not evident. Also, $^{21}\text{Ne}/^{22}\text{Ne}$ ratios fall to the left of the air-solar mixing line in Fig. 2A, although this is a relatively weak constraint because the deviations are only $\sim 1\sigma$.

Isotopic evidence that a He component resembling Q-He is also present would go far toward establishing a Q-gas source. However, as discussed next, the $^3\text{He}/^4\text{He}$ ratios observed in St-1 and St-2 are approximately twice the Q-He ratio. So, the light cometary noble gases, if they have a Q-affinity, appear to be distinct from the meteoritic component in this respect. Their $^4\text{He}/^{20}\text{Ne}$ elemental ratios of ~ 1 to 20 are also distinct, much below the ~ 80 to 140 range in phase Q (Table 1).

Detection of ^3He , which cannot derive from terrestrial contamination, is by itself firm evidence that indigenous gases are indeed present in Stardust samples. The $^3\text{He}/^4\text{He}$ ratio is important because in principle it can point to when comets acquired their noble gases, and from what volatile reservoir—the early protosolar nebula, a locus near the evolving Sun after the deuterium burning that elevated the level of protosolar ^3He , or by later implantation of solar wind radiation. Of the compositions plotted in Fig. 2B, the jovian atmospheric ratio is thought to reflect the primordial nebular (protosolar) value, “D-burning” is an estimate of $^3\text{He}/^4\text{He}$ in the Sun following deuterium burning, and “Solar Wind” represents measurements of the ratio in collectors of the contemporary solar wind returned by the Genesis Mission.

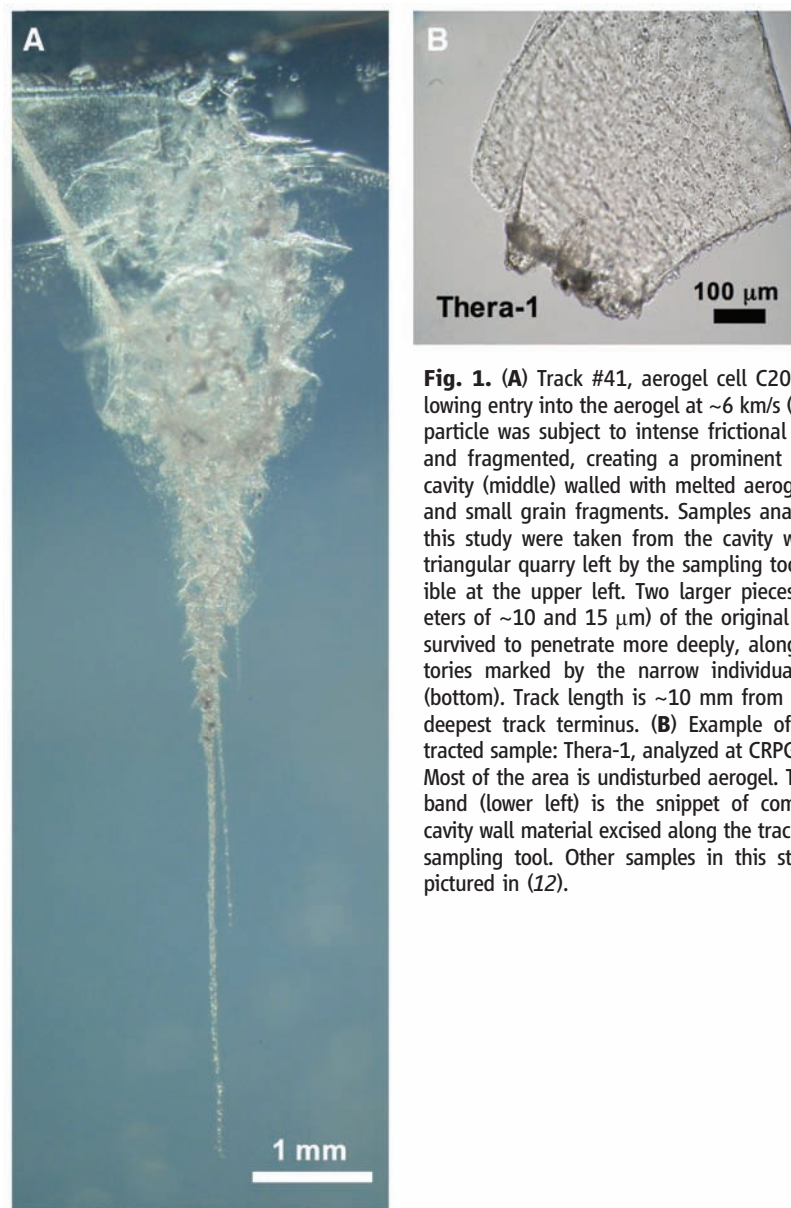


Fig. 1. (A) Track #41, aerogel cell C2044. Following entry into the aerogel at ~ 6 km/s (top) the particle was subject to intense frictional heating and fragmented, creating a prominent bulbous cavity (middle) walled with melted aerogel silica and small grain fragments. Samples analyzed in this study were taken from the cavity wall; the triangular quarry left by the sampling tool is visible at the upper left. Two larger pieces (diameters of ~ 10 and $15 \mu\text{m}$) of the original particle survived to penetrate more deeply, along trajectories marked by the narrow individual tracks (bottom). Track length is ~ 10 mm from entry to deepest track terminus. (B) Example of an extracted sample: Thera-1, analyzed at CRPG Nancy. Most of the area is undisturbed aerogel. The dark band (lower left) is the snippet of compressed cavity wall material excised along the track by the sampling tool. Other samples in this study are pictured in (12).

Table 1. Helium and neon abundances, relative abundances, and isotope ratios in Stardust particle track samples. Uncertainties are $\pm 1\sigma$ (NM: not measured). Limits for ^4He and $^3\text{He}/^4\text{He}$ in St-3 reflect the probable presence of an anomalous ^4He blank contribution (12). Phase Q and Solar compositions are listed for comparison. Phase Q data are from (15). Solar (solar wind) data are averages or ranges of measurements in Genesis collector materials (16–18).

| Sample | ^4He ($\times 10^{-11}$ cm 3 STP) | $^3\text{He}/^4\text{He}$ ($\times 10^{-4}$) | ^{20}Ne ($\times 10^{-11}$ cm 3 STP) | $^{20}\text{Ne}/^{22}\text{Ne}$ | $^{21}\text{Ne}/^{22}\text{Ne}$ ($\times 10^{-2}$) | $^4\text{He}/^{20}\text{Ne}$ |
|----------------------|---|---|--|---------------------------------|---|------------------------------|
| St-1* | 7.27 ± 0.36 | 2.92 ± 0.26 | 5.34 ± 0.42 | 10.68 ± 0.35 | 2.73 ± 0.24 | 1.36 ± 0.13 |
| St-2* | 5.26 ± 0.30 | 2.47 ± 0.34 | 0.94 ± 0.14 | 9.0 ± 1.6 | 3.9 ± 1.1 | 5.60 ± 0.89 |
| St-3* | $<66.9 \pm 2.4$ | $>0.318 \pm 0.032$ | 1.06 ± 0.15 | 9.3 ± 1.6 | 4.2 ± 1.8 | $<63.1 \pm 9.2$ |
| Thera-1 [†] | 53 ± 17 | NM | 2.47 ± 0.65 | 12.9 ± 3.2 | 2.45 ± 0.29 | 21.4 ± 4.9 |
| Thera-2 [†] | 17.5 ± 3.8 | NM | 5.80 ± 0.43 | 10.49 ± 0.24 | 2.79 ± 0.17 | 3.0 ± 1.3 |
| Phase Q | | 1.45 ± 0.15 | | 10.1–10.7 | 2.78–2.94 | 110 ± 30 |
| Solar | | 4.44 ± 0.10 | | 13.90 ± 0.08 | 3.35 ± 0.07 | 520–670 |

*Analyzed at the University of Minnesota, USA.

[†]Analyzed at CRPG Nancy, France.

The differences between the Q-He and observed $^3\text{He}/^4\text{He}$ ratios (Fig. 2B) cannot be attributed to excess ^3He produced by nuclear spallation during space exposure to high-energy galactic cosmic rays (GCRs). Although the abundances of ^3He in these samples are small (<600,000 atoms), concentrations per gram of host grains are not. The mass density of grain fragments lodged in the bulbous cavity wall, assessed by Fe fluorescence measurements, is $\sim 350 \text{ ng/cm}^2$ assuming uniform areal density, and the average area of cavity wall material in each of the five analyzed samples is $\sim 7.3 \times 10^{-4} \text{ cm}^2$ (supporting online text). The average grain mass per sample is then $\sim 0.26 \text{ ng}$, and the corresponding ^3He concentration, obtained by dividing this mass into the Table 1 abundances, is $\sim 7.1 \times 10^{-5} \text{ cm}^3$

STP/g. About half of this would have to be spallogenic to elevate $^3\text{He}/^4\text{He}$ from Q-He to the measured ratio (Fig. 2B). Calculations (supporting online text) show that this would require the particle to reside within the upper meter of the surface, where ^3He production is maximum, for times exceeding the age of the solar system. In reality, abundances of spallation products in the active surface of a comet should be essentially nil; shallow zones where they might conceivably accumulate over long times have eroded away. Average sublimation erosion of the Wild 2 surface just since its 1974 appearance is estimated to be $\sim 1 \text{ m}$ (19), corresponding to $\sim 5 \text{ m}$ in the $\sim 20\%$ of the surface covered by the gas and dust jets encountered by Stardust (20) if these source areas were active on previous apparitions.

With a degassed grain mass of $\sim 0.26 \text{ ng}$, average concentrations of the principal isotopes ^4He and ^{20}Ne (Table 1) are ~ 1 and $\sim 0.1 \text{ cm}^3 \text{ STP/g}$, respectively. These high values have important implications for how the Stardust samples acquired their noble gases. It is clear (Fig. 3) that equilibrium solubility of nebular Ne into melt—from which the Stardust minerals are assumed to have crystallized—is many orders of magnitude too small at calculated nebular pressures to match observed Ne concentrations. A similar shortfall is seen for physical adsorption of ambient Ne in grain-surface structures, even for a favorable case, shown here, of adsorption on finely divided carbon. In contrast, Stardust Ne concentrations are interestingly comparable to those implanted in IDPs and lunar regolith grains by solar-wind ion irradiation (Fig. 3). Among known gas acquisition mechanisms, it appears that only ion implantation could generate such high Ne (and He) loading in these mineral grains.

Fig. 2. (A) Neon isotope ratios in Ne-Q, and in Stardust samples St-1 and Thera-2 (Table 1). Ne-Q isotopic range (shaded area) is from (15). The solid line extending from Air-Ne defines compositions generated by mixtures of air with solar wind Ne, offscale at $^{20}\text{Ne}/^{22}\text{Ne} = 13.90 \pm 0.08$, $^{21}\text{Ne}/^{22}\text{Ne} = 0.0335 \pm 0.0007$ (16–18). (B) Helium isotope ratios in St-1 and St-2 (Table 1), compared to $^3\text{He}/^4\text{He}$ measured in other solar-system reservoirs: $1.45 \pm 0.15 \times 10^{-4}$ in He-Q (15), $1.66 \pm 0.05 \times 10^{-4}$ in Jupiter's atmosphere (29), $4.44 \pm 0.10 \times 10^{-4}$ in Genesis measurements of present-day solar wind (16–18). The ratio in the Sun following deuterium burning is estimated to be $3.65 \pm 0.38 \times 10^{-4}$ (30). Error bars in (A) and (B) are 1σ .

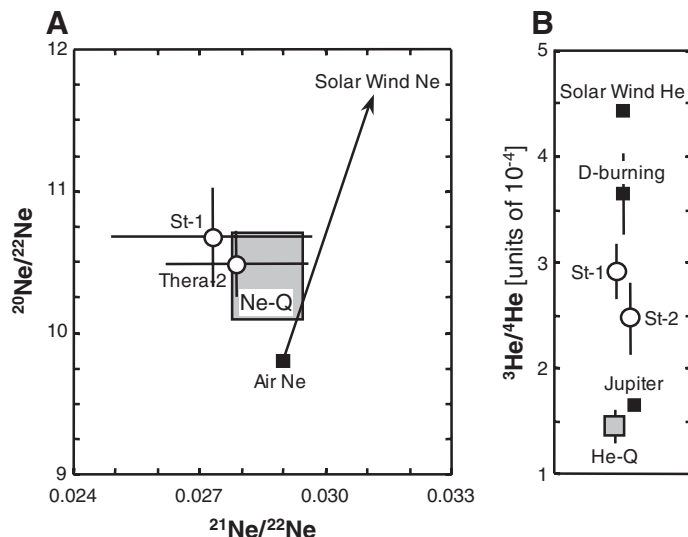
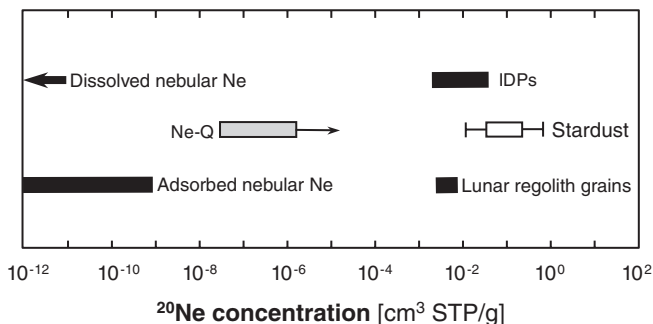


Fig. 3. Concentration ranges of ^{20}Ne in $\text{cm}^3 \text{ STP/g}$ ($1 \text{ cm}^3 \text{ STP} = 2.69 \times 10^{19}$ atoms) in the Stardust samples, the Q-Ne carrier phase (15), ~ 5 - to $10\text{-}\mu\text{m}$ -diameter IDPs (31), and ~ 1 - to $10\text{-}\mu\text{m}$ lunar regolith grains (32) irradiated by the solar wind; measured in the laboratory for adsorption on carbon black at $\sim 20^\circ$ to 400°C (33); and calculated for Ne dissolved in basaltic melt and partitioned into Fe crystals (there are no available data for FeS partitioning), using gas-melt distribution and iron-melt partition coefficients from (34) and (35). Stardust concentrations are from measured ^{20}Ne abundances (Table 1) and average host grain masses of 0.26 ng per sample. The plotted IDP range is for the Group 2 IDPs in (31), which have an average He isotope ratio consistent with implanted solar wind. Concentrations of adsorbed and dissolved Ne depend on Ne partial pressure in the ambient gas. Plotted ranges assume a total nebular pressure of 10^{-3} atm (100 Pa) close to the Sun (36), and a corresponding Ne partial pressure of 10^{-7} atm (37). The highest concentration of dissolved Ne, at $\sim 10^{-11} \text{ cm}^3 \text{ STP/g}$, is for equilibrium solution into basaltic melt (34); the arrow points to concentrations partitioned into iron crystals, lower by a factor of ~ 10 at low pressure (35). Error bars on the Stardust range reflect a conservative estimate of a factor 3 uncertainty in calculations of concentrations.



In the context of a possible Q-gas source for Stardust Ne, one sees in Fig. 3 that the Q-Ne abundances per gram of “carrier phase” in meteorites are lower than the Stardust Ne range by factors of $\sim 10^5$; the corresponding factors for He are $\sim 10^4$. However, this “carrier phase,” a residue isolated by chemical dissolution of silicate minerals, is itself a mixture of several mineralogic and organic components. The actual hosts of Q-gases appear to be organic materials (21, 22), but their masses, though much debated, are essentially unknown (15). It seems likely that the true Q-carriers constitute only a fraction of total residue mass, and thus the Fig. 3 concentrations are indicated as lower limits, possibly by orders of magnitude.

Where do we presently stand on interpreting these Ne and He distributions in Stardust material? There are two important issues. One is the nature of the material carrying the noble gases. We know from the high temperatures ($\geq 1250^\circ\text{C}$) needed to release them that the host is refractory, consistent with the mineralogy of the grains lodged in the track wall that we have assumed to be the carriers. But, just on this basis, they are not the only option; organic carriers of meteoritic Q-gases also retain major fractions of their gases until heated to $>1200^\circ\text{C}$ (23). Some Stardust particle tracks are rich in carbonaceous matter (24), raising the possibility of an organic host. There are indications, however, that this one is not. An IR spectroscopy search of a small sample of bulb-wall material and adjacent aerogel for organics was negative, setting an upper limit of $\sim 10 \text{ fg}$ on average in the samples analyzed for noble gases assuming that organic materials are uniformly distributed in the track wall (supporting online text). Average ^{20}Ne and ^4He concentrations would then have to exceed ~ 3 liters/g and ~ 20 liters/g, respectively, if the gases were trapped in this amount of organic matter. Trapping capacities in organic compound structures could not approach these levels. A caution here, be-

cause the IR search was limited to a small fraction of the total bulb wall area, is that higher abundances of heterogeneously distributed organics could be sited elsewhere in the track and in the samples. However, a large fraction of these organics, if present, is likely to be barren of noble gases. There is evidence that many of the Stardust organic components are labile, volatilized during particle capture and diffused into aerogel adjacent to tracks (24). Helium and Ne released by labile carrier volatilization probably escaped on impact, because we have argued earlier, on the basis of high release temperatures, that Stardust gases are not trapped in aerogel. The most likely organic carriers are therefore those refractory enough to retain their gases during impact heating. What fraction of total organics these might constitute is unknown.

The alternative that the Stardust He and Ne are carried in FeS-Fe grains is consistent with the finding that abundant Q-gases are present in sulfide- and metal-rich separates from certain meteorites (25), although this study did not prove that the gases were actually sited in these phases. We have shown that the required Ne loading of such grains, while large, is not implausible (Fig. 3). Whether this is also the case for a putative organic carrier remains to be seen. Either way, however, the high noble gas concentrations reported here are unlikely to be much lower, no matter what the carrier actually is.

The second issue concerns the compositional nature of the source reservoir. Neon isotopes resemble Q-Ne, but a Q-like source for both Ne and He encounters the problems of the high $^3\text{He}/^4\text{He}$ and low $^4\text{He}/^{20}\text{Ne}$ ratios (Fig. 2B and Table 1). These latter ratios, however, do not necessarily reflect the elemental composition of the source reservoir. Helium thermally escapes from trapping sites more readily than Ne, so low $^4\text{He}/^{20}\text{Ne}$ could result from diffusive loss from grains during gas incorporation or over their metamorphic history. Diffusion also alters isotope ratios, with the lighter species escaping preferentially, and this led a reviewer to ask if the gases initially implanted in the grains could have been solar rather than Q-like—i.e., a direct sampling of the ancient Sun, subsequently fractionated from solar ratios (Table 1) to the observed composition by extreme diffusive losses. An alternative might be gas acquisition from an initially solar reservoir that had been fractionated by mass-dependent losses to space. Both hypotheses, however, fail a central test: Diffusion calculations (supporting online text) show that replication of the observed Ne and He isotope ratios requires the percentage of solar Ne depletion to greatly exceed that of the lighter He, an unphysical result either for diffusion from natural minerals or for Rayleigh distillation of a gaseous reservoir.

We conclude that the Stardust Ne data point more firmly to a Q-gas source than to other alternatives. The anomalously high $^3\text{He}/^4\text{He}$ and low $^4\text{He}/^{20}\text{Ne}$ ratios remain unexplained, although

it may be possible to reconcile them with the Q-like Ne by invoking a multistage model of gas acquisition and loss (supporting online text). The presence of Q-gases would be an important clue to the antiquity of the Wild 2 gas reservoir. They have just the characteristics one would expect for a fundamental constituent of the early solar nebula. They are present in all meteorites that have been subject to the experimental procedures required to isolate their carrier phase (15), and $^3\text{He}/^4\text{He}$ in Q is close to the Jupiter atmospheric ratio (Fig. 2B), a marker for He composition in the primitive nebula.

A central question for the noble gases is where they were acquired by the comet particles. One of the surprises in samples collected from an icy object forming, and, for much of its lifetime, residing in the cold outer reaches of the solar system was the discovery that many of its constituent particles, including that in track 41, are igneous, refractory “rocklets” processed at very high temperatures, presumably close to the Sun before transport to the outer nebula and incorporation into Wild 2 (1, 26–28). Another is the finding here of high concentrations of He and Ne that, of known gas acquisition mechanisms, only ion irradiation seems capable of explaining (Fig. 3). These observations, together with the isotopic data in Fig. 2—pointing to a source containing primitive Q-Ne, and a He composition that could reflect later addition of a solar component to diffused Q-He—suggest that Stardust grains and the carbonaceous carrier now found in meteorites sampled the same Q-gas reservoir, the grains by ion implantation in an energetic environment near the young evolving Sun.

Particles transported from hot inner zones of the solar nebula are thought to constitute ~10% of the mass of Wild 2 (1). The large gas concentrations reported here may therefore have important implications for the supply of comet-carried volatiles to planets if these particles are collectively characterized by similar concentrations and their mass fraction in Wild 2 is typical of comets in general. The isotope ratios and high concentrations of Ne are particularly relevant in efforts to model cometary noble gas contributions to planetary atmospheres (9, 10).

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Supporting Online Material

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Materials and Methods
SOM Text
Figs. S1 to S3
Table S1
References

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