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Constraints on Neon and Argon Isotopic Fractionation in Solar Wind

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crystals, will doubtless reveal other core structures, from which rules or principles of core assembly may ultimately be derived. It remains to investigate the chemical and physical properties of the Au_{102} nanoparticle, as well as to explore the theoretical basis of the gold packing and gold-thiol interactions that we have observed.

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Constraints on Neon and Argon Isotopic Fractionation in Solar Wind

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To evaluate the isotopic composition of the solar nebula from which the planets formed, the relation between isotopes measured in the solar wind and on the Sun's surface needs to be known. The Genesis Discovery mission returned independent samples of three types of solar wind produced by different solar processes that provide a check on possible isotopic variations, or fractionation, between the solar-wind and solar-surface material. At a high level of precision, we observed no significant inter-regime differences in $^{20}Ne/^{22}Ne$ or $^{36}Ar/^{38}Ar$ values. For $^{20}Ne/^{22}Ne$, the difference between low- and high-speed wind components is 0.24 ± 0.37%; for $^{36}Ar/^{38}Ar$, it is 0.11 ± 0.26%. Our measured $^{36}Ar/^{38}Ar$ ratio in the solar wind of 5.501 ± 0.005 is 3.42 ± 0.09% higher than that of the terrestrial atmosphere, which may reflect atmospheric losses early in Earth's history.

Planetary materials formed from a disk of gas and dust around the early Sun, which we refer to as the solar nebula. As a standard model, planetary scientists assume that the elemental abundances and especially the isotopic compositions of elements in the nebula are uniform and that the nebular composition is preserved in the solar outer convective zone (*1*). Thus, allowing for relatively well-understood nuclear and physical/chemical isotope fractionation, terrestrial isotopic compositions should be the same as in other solar-system materials. To very high precision, this appears to be true for nonvolatile elements (*1*). However, the standard model fails for the isotopes of O, H, N, and the noble gases where large variations (compared to nonvolatile elements) are observed among terrestrial, lunar, meteoritic (asteroidal), and martian materials (*2*–*4*). Because of the nuclear conversion of D to ^{3}He, solar H is monoisotopic, and $^{3}He/^{4}He$ is greatly enhanced. Despite these exceptions, the surface layers of the Sun should preserve the nebular isotopic compositions of C, N, O, and the noble gases (*5*, *6*).

Plasma flowing from the Sun as the solar wind permits the sampling of solar matter. The Apollo Solar Wind Composition (SWC) experiment (*7*) measured relatively precise He, Ne, and Ar isotopic compositions for 1–to–3-day periods in 1969–1972. Here, we address whether the isotopic compositions of Ne and Ar, measured in the solar wind, have changed ("fractionated") from those measured on the surface of the Sun. Ulysses and Advanced Composition Explorer (ACE) spacecraft data have shown that relative proportions of elements in the solar wind are fractionated by amounts correlated with the elemental first ionization potential (FIP) (*8*). FIP fractionation presumably arises because of the preferential extraction of ions relative to atoms during transport into the solar corona from lower levels (*9*). Although the FIP is an atomic property, FIP fractionation models (*9*) predict some isotope effects, but in many specific models, these effects are small. The acceleration of heavier elements from the solar corona into the solar wind can be due to their collisions with protons ("coulomb drag"), and if the drag is incomplete,

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heavy-element density enhancements result (*10*, *11*). These enhancements depend on ion mass as well as charge and would fractionate isotopes.

Genesis exposed materials to the solar wind for 27 months (December 2001 to April 2004) (*12*). Separate samples of the three types ("regimes") of solar wind were collected. Regime-specific collector arrays were deployed according to the flow regime that was present, as determined by on-board solar-wind monitors (*13*). These regimes were: (i) low-speed, mostly less than 475 km/s but allowing for hysteresis during transitions (*13*); (ii) high-speed, onset generally 525 km/s; and (iii) coronal mass ejection (CME). CMEs are spectacular bubbles of plasma erupted in discrete events. We use the shorthand labels (i) L, (ii) H, and (iii) CME for these regimes, in addition to bulk solar wind. The quality of the regime separation has been verified by comparison with data from other spacecraft for the Genesis collection period; for instance, the H regime sample is verified to be composed of coronal-hole material by association with a low O^{7+}/O^{6+} ratio using O charge-state data from the ACE/Solar Wind Ion Composition Spectrometer instrument (*14*).

Genesis carried different collectors that were optimized for specific elements. Here, we report Ne and Ar results from aluminum-deposited-on-sapphire (AloS) collectors. The low atomic number of Al minimizes backscatter. AloS has low blanks and high noble-gas retention (*15*, *16*). AloS has probably suffered some He loss; therefore, we report only Ne and Ar results.

An infrared (IR)-laser–extraction technique was specifically designed (*17*), as it selectively evaporates only the Al film on the IR-transparent sapphire substrate. Because the laser interacts with only Al and does not impinge upon the sample-system walls, laser extraction of AloS has low blanks. Further experimental details are available in the supporting online material (*16*).

When compared with data from the Apollo SWC foils (*7*), our findings for Ne and Ar isotopic and elemental ratios validate these results but are significantly more precise (Table 1, fig. S1, and tables S1 and S2). Small (1%) but statistically significant interlaboratory variations exist between reported bulk Genesis $^{20}Ne/^{22}Ne$ values (*18*, *19*), but for assessing inter-regime differences, the precision of the data from one laboratory and instrument is what is important. To a high degree of precision, our data show no measurable differences in the $^{20}Ne/^{22}Ne$ ratio among different regime samples.

^{40}Ar is rare in solar composition but dominates atmospheric Ar because of the strong depletion of noble gases in the formation of Earth and the subsequent production from ^{40}K decay in terrestrial rocks. The measured ^{40}Ar composition (table S2) can be accounted for by system blanks with no clear contribution from particulate contamination resulting from the crash of the recovery capsule. At the 2σ level, there are no variations in $^{36}Ar/^{38}Ar$ values among the different solar-wind regime samples (Table 1). CME $^{36}Ar/^{38}Ar$ is slightly heavier than the others at 1σ but the uncertainty for this regime is almost twice as large, suggesting statistical variations. The difference between the L and H regimes for $^{36}Ar/^{38}Ar$ is 0.11 ± 0.26% (less than 1σ). Our bulk samples define a precise solar-wind $^{36}Ar/^{38}Ar$ composition of 5.501 ± 0.005.

The highest scientific objectives for Genesis are to measure the solar isotopic compositions of N and O, for which there are unexplained variations among planetary materials. Even small isotopic fractionations between the solar wind and the solar surface are of major importance for planetary sciences. Genesis independently sampled three regimes formed by different solar processes. If isotopic fractionation occurs, it is plausible that the amounts would vary among the regimes; thus the comparison of regime Ne and Ar isotopic compositions will test for fractionations. Specifically, coulomb drag effects (*11*) predict that fractionations depend directly on mass and indirectly on speed; consequently, a simple, measured quantity to assess solar-wind isotopic fractionation is the fractional difference between an L and an H regime sample. Qualitatively, light-isotope enrichment (for example, enrichment in $^{20}Ne/^{22}Ne$ values from L regime samples, relative to those from H regime samples) is predicted. The fractionation should be larger for $^{20}Ne/^{22}Ne$ than for $^{36}Ar/^{38}Ar$ because of a larger relative-mass difference. The solar He/H ratio is double that of the solar wind (*20*). If coulomb drag is a major cause of the He/H difference, then solar/solar-wind fractionation may be significantly greater than the differences between regimes.

We found no significant variations in the isotopic compositions of Ne and Ar at the 1σ level. For $^{20}Ne/^{22}Ne$, the L versus H regime difference is 0.24 ± 0.37%, corresponding to an upper limit of 0.98% at the 2σ level. Qualitatively similar limits would apply for O and N. For $^{36}Ar/^{38}Ar$, the L versus H regime difference is 0.11 ± 0.26%, corresponding to a 2σ upper limit of 0.63%.

Unlike isotopes, elemental ratios among the different regimes are not expected to be constant. Because of systematic calibration uncertainties, only relative $^{20}Ne/^{36}Ar$ results are interpretable. The $^{20}Ne/^{36}Ar$ errors (precision) (Table 1) are estimated to be about 9%, on the basis of replicate analyses of the ^{20}Ne and ^{36}Ar fluxes in the bulk sample (tables S1 and S2). At the 1σ level, $^{20}Ne/^{36}Ar$ ratios for the CME and H regime samples are the same, but L regime samples differ from the other two by about 25%. Ne has a higher FIP than Ar, so even though the solar abundances of Ne and Ar are unknown, the lower $^{20}Ne/^{36}Ar$ value for L samples indicates a greater FIP fractionation for the low-speed solar wind. This agrees with spacecraft data based on O/Si ratios (*21*). The Apollo SWC $^{20}Ne/^{36}Ar$ value is closest to our L regime value, and most of the Apollo SWC collection occurred under low-speed conditions (*7*).

A standard approach to obtaining solar abundances is to extrapolate correlation plots of solar-wind compositional variations to some known solar value (*6*). In our case, there are no variations to extrapolate; thus, our isotopic data support the previously made association of solar-wind and solar-nebula Ne and Ar isotopic compositions (*22*).

As early as the 1960s, meteoritic data on noble-gas isotopic compositions challenged the standard model of an isotopically homogeneous solar nebula. Today these data are partially understood in terms of the large contributions to bulk meteorite noble-gas concentrations by extant presolar grains (*4*). However, presolar grains cannot account for the observed O and N isotopic variations among planetary materials (*2*, *4*) and for the relatively large amounts of isotopically distinct noble gases, variously referred to as "planetary" or "Q" (*22*).

The Apollo SWC (*7*) experiment showed that the solar-wind $^{20}Ne/^{22}Ne$ ratio was highly enriched compared to the terrestrial atmospheric ratio (Table 1). This proved that volatile-element

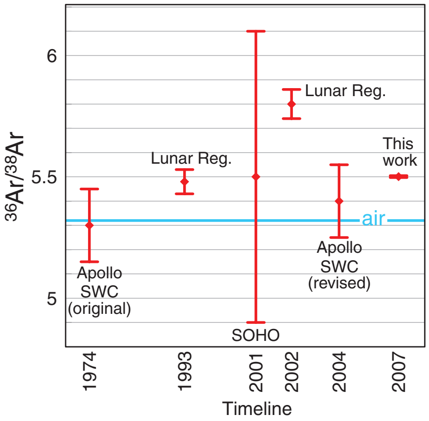

Fig. 1. Comparison of solar-wind Ar measurements: Apollo SWC foils (*7*, *28*), lunar regolith (Reg.) (*26*, *27*), SOHO (*25*), and terrestrial atmosphere (*29*).

Table 1. Ne and Ar isotopic and elemental ratios from AloS collectors. Ne is corrected for backscattering. Errors are 1σ.

Sample, collection time	$^{20}Ne/^{22}Ne$	$^{36}Ar/^{38}Ar$	$^{20}Ne/^{36}Ar$
Bulk, 852.83 days	13.972 ± 0.025	5.501 ± 0.005	59 (5)*
High-speed, 313.01 days	13.956 ± 0.041	5.502 ± 0.010	66 (6)*
CME, 193.25 days	13.979 ± 0.031	5.467 ± 0.017	59 (5)*
Low-speed, 333.67 days	13.990 ± 0.031	5.508 ± 0.010	46 (4)*
Apollo SWC (*7*)	13.7 ± 0.15	5.4 ± 0.15	49 ± 7
Terrestrial atmosphere (*28*)	9.80 ± 0.08	5.319 ± 0.011	0.524 ± 0.002
Q (*22*)	13.7	5.33	0.049

*Numbers in parentheses indicate 9% estimated errors based on the scatter in measured amounts of ^{20}Ne and ^{36}Ar.

isotopic compositions varied among planetary materials. For ^{20}Ne/^{22}Ne, the 38% difference may reflect isotopic fractionation accompanying an early loss of the terrestrial atmosphere (*23*), an extreme example of chemical/physical mass-dependent isotope fractionation. However, the report by Clayton in 1972 of "non–mass-dependent" O isotope fractionations (*24*) clearly showed that isotopic variations in volatile elements reflect isotopic inhomogeneities in the solar nebula, an unequivocal failure of the standard model.

A major problem that remains is that of understanding the differences between solar-wind isotopic noble-gas abundances and those of meteoritic (Q) noble gases (*22*) (Table 1). These abundances should have been similar if, as expected, Q noble gases are samples of the solar nebula trapped in meteoritic material. Can a single mass-dependent fractionation process simultaneously explain large differences in ^{20}Ne/^{22}Ne and small but significant differences in other noble-gas isotope ratios, such as ^{36}Ar/^{38}Ar between Q and solar-wind noble gases? Although fractionation during adsorption onto grain surfaces has often been discussed, the fractionation process is basically unknown.

Unlike those of Ne, Ar isotopic variations, although widely believed to exist, were not quantitatively well-defined. Isotopic ratios from spacecraft instruments such as the International Solar and Heliospheric Observatory (SOHO)/mass time-of-flight spectrometer (*25*) (Fig. 1) have insufficient precision to address planetary science issues (e.g., differences between solar-wind and terrestrial ^{36}Ar/^{38}Ar ratios). The short Apollo SWC exposure did not yield a sufficiently precise ^{36}Ar/^{38}Ar value to distinguish the solar and atmospheric ratios (*7*). A higher-than-terrestrial solar ^{36}Ar/^{38}Ar ratio is safely inferred from studies of lunar soils, but there has been debate on how much higher it can be, with ^{36}Ar/^{38}Ar values reported between 5.48 and 5.80 (*26*, *27*). Corrections for excess ^{38}Ar from galactic–cosmic-ray nuclear reactions present a challenge in interpreting lunar-soil data. Genesis data require negligible galactic–cosmic-ray corrections. This, plus higher analytical precision, significantly improves the accuracy of the solar-wind and the atmospheric ^{36}Ar/^{38}Ar difference. Our solar-wind ^{36}Ar/^{38}Ar ratio (Table 1) is higher than that of the terrestrial atmosphere by 3.42 ± 0.09%. This should lead to improved constraints on models for terrestrial atmospheric loss. It is also significantly higher than the ^{36}Ar/^{38}Ar value of "Q".

The significantly increased precision of Ne and Ar solar-wind isotopic compositions have yielded no differences among the Genesis regime samples. Nevertheless, we have not proved that there is no Ne or Ar isotopic fractionation between the Sun and the solar wind. Our data clearly put a constraint on such fractionations, but at present the strength of the constraint is not known. A "unified" theoretical approach evaluating FIP and coulomb drag fractionations in the context of formation models of the different solar-wind regimes is required. If fractionations between the Sun and the solar wind are large but the same for all regimes, then the absence of inter-regime isotopic variations is inconclusive. Alternatively, the fractionations among regimes could be large and the lack of inter-regime variations quite constraining. The goal of the Genesis mission is to provide higher-precision solar-wind composition data, leading to better theories of solar-wind fractionations, which, in turn, will lead to improved solar abundances for planetary science purposes.

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Southern Hemisphere and Deep-Sea Warming Led Deglacial Atmospheric CO_2 Rise and Tropical Warming

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Establishing what caused Earth's largest climatic changes in the past requires a precise knowledge of both the forcing and the regional responses. We determined the chronology of high- and low-latitude climate change at the last glacial termination by radiocarbon dating benthic and planktonic foraminiferal stable isotope and magnesium/calcium records from a marine core collected in the western tropical Pacific. Deep-sea temperatures warmed by ~2°C between 19 and 17 thousand years before the present (ky B.P.), leading the rise in atmospheric CO_2 and tropical–surface-ocean warming by ~1000 years. The cause of this deglacial deep-water warming does not lie within the tropics, nor can its early onset between 19 and 17 ky B.P. be attributed to CO_2 forcing. Increasing austral-spring insolation combined with sea-ice albedo feedbacks appear to be the key factors responsible for this warming.

The data obtained from high-latitude ice cores establish a close temporal relation between varying concentrations of atmospheric CO_2 and atmospheric temperatures during glacial terminations (*1*). However, uncertainty in the gas-age chronologies and inadequate temporal resolution in many proxy climate reconstructions have hampered efforts to establish the exact phasing of events during glacial terminations, a necessary step in understanding the physical relation between CO_2 forcing and climate change (*2*). Arguably, the most robust estimate of changes in mean global temperatures that accompany glacial terminations is the amount of heat stored in the oceans. The large size of the oceanic reservoir and the long residence time of