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A $^{15}\text{N}$-Poor Isotopic Composition for the Solar System As Shown by Genesis Solar Wind Samples

B. Marty, M. Chaussidon, R. C. Wiens, A. J. G. Jurewicz, D. S. Burnett

The Genesis mission sampled solar wind ions to document the elemental and isotopic compositions of the Sun and, by inference, of the protosolar nebula. Nitrogen was a key target element because the extent and origin of its isotopic variations in solar system materials remain unknown. Isotopic analysis of a Genesis Solar Wind Concentrator target material shows that implanted solar wind nitrogen has a $^{15}\text{N}/^{14}\text{N}$ ratio of 2.18 ± 0.02 × 10^{-3} (that is, ≈40% poorer in $^{15}\text{N}$ relative to terrestrial atmosphere). The $^{15}\text{N}/^{14}\text{N}$ ratio of the protosolar nebula was 2.27 ± 0.03 × 10^{-3}, which is the lowest $^{15}\text{N}/^{14}\text{N}$ ratio known for solar system objects. This result demonstrates the extreme nitrogen isotopic heterogeneity of the nascent solar system and accounts for the $^{15}\text{N}$-depleted components observed in solar system reservoirs.

Nitrogen, the fifth most abundant element in the Sun, is particularly intriguing because its isotopic composition shows variations up to a factor of ≈6 in solar system objects (excluding presolar grains), either at a microscopic scale or at the planetary scale (2, 6–9). Over the past few decades, numerous attempts have been made to determine the N isotopic composition of the Sun through the analysis of solar ions implanted in lunar soils; however, both $^{15}\text{N}$-rich and $^{15}\text{N}$-poor components were found on minerals from lunar soils (8, 9). These variations were first thought to represent a secular change of the nitrogen isotopic composition of the SW (9), but no known solar process could be invoked to produce these variations, and mixing between solar and planetary N was later advocated (7, 10). More recently, ion-probe depth profiling of single lunar grains demonstrated that at a depth of ≈50 nm, solar H (D-free) was associated with $^{15}\text{N}$-poor N (e.g., depleted by at least ≈24% relative to terrestrial) and, thus, presumably of solar origin (6).

**Ion-probe analysis of Genesis Concentrator silicon carbide target.** We report here the precise isotopic analysis of SW N collected by implantation into a silicon carbide target (SIC) quadrupole of the target of the Genesis Solar Wind Concentrator, an electrostatic mirror (11) that increased the fluence of some SW elements by a factor up to 50 (12, 13). We analyzed along a traverse of the quadrant by secondary ion mass spectrometry using the Cameca 1280HR instrument recently installed at CRPG Nancy, France (14). This instrument has improved transfer optics and sensitivity of primary and secondary beams that permits very-high-mass (up to $m/\Delta m = 300,000$) and high-precision isotopic ratio measurements in multicollection mode. The SIC target was sputtered using a 10-kV Cs primary beam, and the resulting secondary $^{12}\text{C}^{14}\text{N}^-$ and $^{12}\text{C}^{15}\text{N}^-$ ions were accelerated at 10 kV (total impact energy of 20 kV) and analyzed in multicollection electron multiplier mode at a mass resolution of ≈8000, so that all potential isotopic interferences were resolved (14). $^{28}\text{Si}$ ions were counted simultaneously to monitor the stability of the secondary beam with time (in a given depth profile), as well as to provide a way to normalize N isotopic counts between different runs. Before analysis, four areas of 100 by 100 μm were cleaned of surface contamination using a low-energy ion beam (14). In each of the four cleaned areas, 6 to 10 measurements were taken by depth profiling at different locations (≈10-15 μm diameter each). Procedural blanks were estimated by analyzing for N isotopes flight-spares SIC target material (kept on Earth) using the same analytical conditions and were found to be negligible (<1%) for most analyses (Fig. 1). The implantation depths and fluorences of SW nitrogen ions were estimated from the analysis of SiC implanted with $^{15}\text{N}$ ions at known energy and fluence (14). Ion-probe instrumental mass discrimination was determined to be ≈29 ± 8.6 per mil (%o) by analyzing an SiC standard under the same conditions and comparing those data with the results previously obtained by laser ablation–static mass spectrometry (14).

Both $^{14}\text{N}$ and $^{15}\text{N}$ data define simple bell-shaped distributions as a function of depth that peak ~80 nm below the target’s surface (Fig. 1), as expected for SW implantation at energies imposed by SW ion velocities and the Concentrator’s ion acceleration (14). The measured $^{15}\text{N}/^{14}\text{N}$ ratio must be corrected for the instrumental mass fractionation (IMF) of the Genesis Concentrator, which varies as a function of position along its radius. This IMF was calibrated previously for the same SIC quadrant as a function of distance from the Concentrator’s center using Ne (that is, by observing the deviation of the Ne isotopic composition extracted by laser ablation from known SW compositions (15). From charge/mass considerations, the fractionation per mass unit of N (and O) isotopes should be comparable to that of Ne isotopes (11, 15); this relation allows the $^{15}\text{N}/^{14}\text{N}$ ratio to be corrected for the Concentrator’s IMF on the basis of the Ne isotope fractionation. The Ne isotopic ratio measured in SiC at a distance of 18 to 20 mm from the Concentrator’s center is not fractionated relative to the bulk SW neon isotopic composition (15, 16). Thus, we assume safely that the N isotopic composition of areas at 19-mm distance is that of the nonfractionated SW nitrogen. For other areas at 11 and 9 mm from the center, we use the Ne isotopic data to correct for Concentrator’s isotopic fractionation (Table 1). However, these corrections are small—on the order of a few per mil (maximum: 19‰)—and mostly within stated errors. All detailed results, after corrections for the IMFs of both the Concentrator and the ion probe, are given in Table 1.

**Isotopic composition of solar wind nitrogen.** Our measurements yield a $^{15}\text{N}/^{14}\text{N}$ ratio for the SW of 2.178 ± 0.024 × 10^{-3} (95% confidence level), corresponding to δ$^{15}\text{N} = -407$ ± 7‰, where

\[
\delta^{15}\text{N} = \left( \frac{^{15}\text{N}}{^{14}\text{N}} \right)_{\text{sample}} - 1 \times 1000 \text{ and ATM refers to the isotope composition of atmospheric nitrogen ([(^{15}\text{N}/^{14}\text{N})_{\text{ATM}} = 3.676 × 10^{-3}].\) By its high precision, this result definitively settles the debate on the N isotopic composition of SW (17–19); it is extremely $^{15}\text{N}$-poor, as proposed in (18, 19). Such a $^{15}\text{N}$-depleted component (the $^{15}\text{N}/^{14}\text{N}$ ratio not being precisely known) was previously suspected to exist in meteoritic matter (2, 20) and the jovian atmosphere (21), but it was not understood as representing the solar composition.

The Sun’s bulk $^{15}\text{N}/^{14}\text{N}$ ratio can be obtained from our Genesis SW measurement after correction for isotopic fractionation taking place during acceleration of SW from the photosphere and, possibly, in the convective zone of the Sun. The effect of solar diffusive element settling plus radiative levitation (DESRL) (22) is estimated to
nucleosynthesis of nitrogen isotopes in solar flares might also have changed the $^{15}\text{N}/^{14}\text{N}$ ratio of the outer Sun. However, the effect is predicted to be negligible in the case of spallation for N, though it might be detectable for Li (32).

Because the PSN is the most $^{15}\text{N}$-depleted, as well as the most gas-rich, reservoir in the solar system, we propose that the N isotope variations among solar system bodies result from variable mixing between a $^{15}\text{N}$-poor gaseous component and solids rich in nitrogen-15. Observed H, N, and O isotopic variations are consistent with variable mixtures of a PSN component with components rich in heavy and rare isotopes (D, $^{15}\text{N}$, and $^{17,18}\text{O}$, respectively) (Fig. 2). Alternatively, these enrichments might have resulted from interactions between photons and matter (e.g., photochemistry, ion-molecule reactions) that took place before or during formation of the solar system (33–36). The existence of a common origin for these strong D-, $^{15}\text{N}$-, $^{17,18}\text{O}$-, and $^{16}\text{O}$-enrichments is a key question.

**Planetary implications.** The agreement of the Sun’s and Jupiter’s outer atmosphere $^{15}\text{N}/^{14}\text{N}$ is of considerable importance, because Jupiter’s atmosphere is enriched in N/H (along with Ar, Kr, Xe, C, and S) by about a factor of 3 compared with the solar photospheric elemental ratios (37, 38). We assume that the N isotopic composition of Jupiter’s atmosphere is representative of the whole planet. These enhancements are usually interpreted as indicating that Jupiter is a mixture of solar nebula gas (the source of H and He) and outer solar system planetesimals [the source of the other, less volatile, elements (37)]. If this interpretation is correct, then only about one-fourth of the N in Jupiter is of nebula origin. Nevertheless, Jupiter has preserved
the solar $^{15}\text{N}/^{14}\text{N}$ ratio, requiring that the N in the planetesimal contribution had a low $^{15}\text{N}/^{14}\text{N}$ ratio, with only very small contributions from the very high $^{15}\text{N}/^{14}\text{N}$ ratio observed in cometary HCN and CN (39) or even the more modest $^{15}\text{N}/^{14}\text{N}$ ratio enrichments measured in bulk inner solar system carbonaceous chondrite material. Thus, either the model for the origin of jovian volatiles is not correct, or the cometary HCN-CN $^{15}\text{N}/^{14}\text{N}$ may not be representative of outer solar system, possibly even cometary, matter.

The extremely $^{15}\text{N}$-poor composition for the PSN provides the $^{15}\text{N}$-depleted component required to account for the N isotope variations previously recognized in solar system objects (Fig. 2). Presolar phases, which show a range of $^{15}\text{N}/^{14}\text{N}$ ratios from $\approx 50$ to $\approx 20,000$ (40), are usually considered to be negligible to the solar system N budget. The average abundances in chondrites of presolar diamonds, SiC, and graphite are $\approx 1000$, $\approx 10$ and $\approx 1$ parts per million (ppm), respectively (40), and their N contents (with the exception of Si$_3$N$_4$) range from $\approx 500$ to $\approx 10,000$ ppm (41). Contrary to unequivocal presolar grains, the origin of meteoritic nanodiamonds is unclear because, although they can contain presolar xenon (42), their C isotopic composition is solar (43). The nanodiamond $^{15}\text{N}/^{14}\text{N}$ ratio $[2.40 \pm 0.03 \times 10^{-3}$ (43)] is similar to that of the PSN, which might also point to a solar origin. It is possible that most nanodiamonds formed in the solar system, with only a small fraction (those hosting isotopically anomalous Xe) being inherited from other stellar systems. In detail, nanodiamonds, which comprise several populations, are enriched in $^{15}\text{N}$ by $58 \pm 17\%$ relative to the PSN composition. Hence, they might have sampled regions of the disk enriched in $^{15}\text{N}$ by addition of a minor, presumably nucleosynthetic, component, possibly the one hosting the anomalous Xe.

Our result also has implications for the origin of volatile elements in terrestrial planets. These elements have similar N and H isotopic compositions to those of the sources of some chondritic matter (Fig. 2). Mixing between a D-rich, $^{15}\text{N}$-rich component, as observed in some comets with a solar reservoir (Fig. 2), cannot account for the relative homogeneity (ignoring factors of 1.5 or less) of N and H isotopic ratios of inner planets and meteorites. Instead, this homogeneity suggests relatively efficient stirring and mixing of at least three (N, H) components in the inner solar system.

Nitrogen isotopic variations in meteorites provide a new cosmochemical tracer for understanding chemical and thermodynamical heterogeneities during condensation, dust aggregation and coalescence, and parent-body processing. Variations of the bulk N isotopic compositions of meteorites (44–46) are now consistent with mixing between a $^{15}\text{N}$-rich end-member, hosted by organics and dominating the carbonaceous chondrite inventory, and a $^{15}\text{N}$-depleted component derived from the PSN, preferentially hosted by silicate and metal phases. Insoluble organic matter in meteorites can be extremely rich in $^{15}\text{N}$ on a micron scale (47, 48) and was probably the source of the large $^{15}\text{N}$ enrichments observed in CR/CB meteorites (49, 50).

The incorporation of PSN nitrogen could have taken place either during nebular condensation or by solution in molten silicates and could have been most efficient under reducing conditions imposed by the composition of the PSN. From the composition of the PSN and adopting canonical conditions for the inner solar system $[\text{pressure } P = 10^{-3} \text{ atm, temperature } T = 1500 \text{ K}, \log f_{O_2} \approx -19$ (where $f_{O_2}$ is the oxygen fugacity) (51)$, $\approx 4$ to 100 ppm of solar N could have been dissolved from the PSN into a basaltic melt [the only composition for which the N solubility as a function of $f_{O_2}$ has been measured (52)], making a substantial fraction of meteorite nitrogen (range: 1 to 1000 ppm N). For the same $P$-T conditions and at $\log f_{O_2} \approx -20.3$ [for enstatite chondrite

### Table 1. Results from the analysis of different areas along the Concentrator’s SiC quadrant (SOM, Figs. 55 to 58).

<table>
<thead>
<tr>
<th>Spot #</th>
<th>$^{15}\text{N}/^{14}\text{N}$ measured</th>
<th>95% CI</th>
<th>$^{20}\text{Ne}^{22}\text{Ne}$ measured</th>
<th>95% CI</th>
<th>$^{15}\text{N}/^{14}\text{N}$ cor. Concentrator’s fractionation</th>
<th>95% CI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Area 1</td>
<td>4</td>
<td>$2.233 \times 10^{-3}$</td>
<td>$2.40 \times 10^{-4}$</td>
<td>13.78</td>
<td>0.04</td>
<td>$2.143 \times 10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>$2.180 \times 10^{-3}$</td>
<td>$2.27 \times 10^{-4}$</td>
<td>13.78</td>
<td>0.04</td>
<td>$2.116 \times 10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>$2.093 \times 10^{-3}$</td>
<td>$2.45 \times 10^{-4}$</td>
<td>13.48</td>
<td>0.08</td>
<td>$2.123 \times 10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>$2.168 \times 10^{-3}$</td>
<td>$3.17 \times 10^{-4}$</td>
<td>13.26</td>
<td>0.06</td>
<td>$2.158 \times 10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>$2.009 \times 10^{-3}$</td>
<td>$2.52 \times 10^{-4}$</td>
<td>2.129</td>
<td>0.05</td>
<td>$2.116 \times 10^{-3}$</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>$2.195 \times 10^{-3}$</td>
<td>$2.61 \times 10^{-4}$</td>
<td>19</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>$2.129 \times 10^{-3}$</td>
<td>$2.05 \times 10^{-4}$</td>
<td>19</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td>$2.143 \times 10^{-3}$</td>
<td>$9.00 \times 10^{-5}$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Note: The measured $^{15}\text{N}/^{14}\text{N}$ ratios and $^{20}\text{Ne}^{22}\text{Ne}$ isotope ratios have been corrected for the instrumental mass fractionation using the Ne isotopic ratios for the corresponding areas. The weighted average of all areas’ data are given at the bottom right of the lower section of the table. These values are finally corrected for the instrumental mass fractionation in the ion probe (corr. IMF), determined from the analysis of standard SiC. ATM refers to the nitrogen isotopic composition of the terrestrial atmosphere.
found in the Isheyevo CH/CA chondrite (47) and with the matrix of the carbonaceous chondrite Acfer 094 which has the most extreme O isotope composition found in meteorites and is thought to be a remnant of early solar system water (53). (Inset) Same diagram with an expanded scale.

References and Notes