THE LOWEST ^{15}N/^{14}N END-MEMBER OF THE SOLAR SYSTEM IS THE SUN.

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Introduction: Nitrogen, the fifth most abundant element in the universe, displays the largest stable isotope variations in solar system reservoirs after hydrogen. Yet the isotopic composition of solar nitrogen, presumably the best proxy of the protosolar nebula composition, is not precisely known. The Genesis spacecraft sampled SW ions during 27 months at the Lagrange point L1. Despite a hard landing of the sample return capsule on Earth, target material could be recovered. Previous attempts to measure the N isotopic composition of N implanted in Genesis collectors gave contrasting results. In one study, N was extracted from a Genesis target (gold layer over a sapphire substrate) by amalgamation of gold with mercury vapour under vacuum [1], and its analysis led the authors to propose that solar N was enriched by ~+300 % in ^{15}N relative to the Earth. In another study [2], nitrogen was extracted together with noble gases by laser ablation from a stainless steel frame covered with a gold film holding targets from the Genesis Concentrator (see below), purified sequentially and analysed by static mass spectrometry. Despite extensive terrestrial N contamination of the analyzed material (which was not designed to be analyzed), it was concluded that SW N is depleted in ^{15}N by 40 (±19, 2σ) % relative to terrestrial N [2]. This was corroborated by a MegaSIMS analysis of Concentrator SiC indicating nearly a factor of 2 depletion in solar-wind ^{15}N relative to terrestrial [3]. We report in the following the precise isotopic analysis of SW N implanted in the Genesis SiC quadrant mounted at the centre of the Solar Wind Concentrator (Fig 1), an electrostatic mirror [4] that increased the solar wind fluence by a factor of ~50 [5].

Experimental: The Genesis SW collector was analyzed by secondary ion mass spectrometry, using the Cameca 1280HR2 ion probe recently installed in CRPG-CNRS, Nancy, France. The SiC quadrant was degassed for several weeks under a pressure of 2×10^{-10} Torr. Analyses were made using a 10 kV Cs$^+$ primary beam of ≈1 nA intensity and ≈ 10 µm size with no raster. Secondary ions were accelerated at -10 kV (so that the impact energy was of 20 kV). Nitrogen isotopes were analyzed as ^{12}C^{14}N$^-$, ^{12}C^{15}N$^-$ in multicollection mode on two electron multipliers (trolley L1 for mass 26, central EM for mass 27). ^{30}Si$^-$ (trolley H1 equipped with a Faraday cup) was counted simultaneously to monitor the stability of secondary ion intensities. For each cycle, ^{12}C^{14}N$^-$, ^{12}C^{15}N$^-$ and ^{30}Si$^-$ intensities were counted during 5 sec each, and 80-100 cycles were carried out for one spot measurement. Measurements were made at a mass resolution M/ΔM of approx. 8,000 that allowed the resolution of major isobaric interferences (e.g. M/ΔM of 7,142 required to resolve ^{13}C$_2$ from ^{12}C^{14}N at mass 26). In such conditions, no correction for isobaric interferences were required for the CN isotopomers. Ion probe instrumental mass fractionation was determined to be -29.5±8.6 ‰,(i.e fractionation favoring the heavy isotope) from the analysis in the same conditions of a SiC standard analysed previously for N isotopes by laser ablation - static mass spectrometry. A SiC target implanted with 2x10^{14} ions of ^{15}N at 33 KeV was used to calibrate the implantation depth and the SW fluence of the flight SiC spots. Four different areas were analyzed along the SiC quadrant radius, located at 19 mm (areas 1 & 2), 11 mm (area 3) and 7 mm (area 4) from the center of the concentrator. Cleaning of the sample surface was done before each analysis by sputtering the area to be analysed with a Cs$^+$ beam of 10 nA accelerated at 5 KV. The depth of ablation due to cleaning was monitored and optimized using the SiC implant. Then between 6 and 10 spots were analyzed within the cleaned surface.

Results: Both ^{14}N and ^{15}N data define simple bell-shaped distributions as a function of depth peaking around 100 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. For comparison are also given blank data (green symbols) obtained from the analysis of a non flight SiC target material, showing that the terrestrial blank was always negligible.

The measured ^{15}N/^{14}N ratio was corrected for mass discrimination induced by the Genesis Concentrator which fractionates abundances and isotopes as a function of position along its radius. This discrimination was calibrated previously for the same SiC quadrant, as a function of distance from the Concentrator's center, from the observed differences in Ne isotopic composition between Ne extracted by laser ablation and known SW compositions [5]. From charge/mass considerations, the fractionation per mass unit of N (and O) isotopes should be comparable to that of Ne isotopes [2,4], so that the ^{15}N/^{14}N ratio can be corrected for Concentrator's discrimination using Ne isotope data. The Ne isotopic ratio measured in SiC at a distance of 18-20 mm from the concentrator's center is not fractionated relative to the bulk SW neon isotopic composition (6), the latter being known from measurement of non-concentrated Genesis collectors. Thus we assume safely that the N isotopic composition of areas at 19 mm distance (areas 1 & 2) is that of the non-fractionated SW nitrogen. For other areas at 11 and 7 mm from the center, we use the Ne isotopic data to correct for concentrator's isotopic fractionation. In both cases corrections are of the same magnitude as the errors.

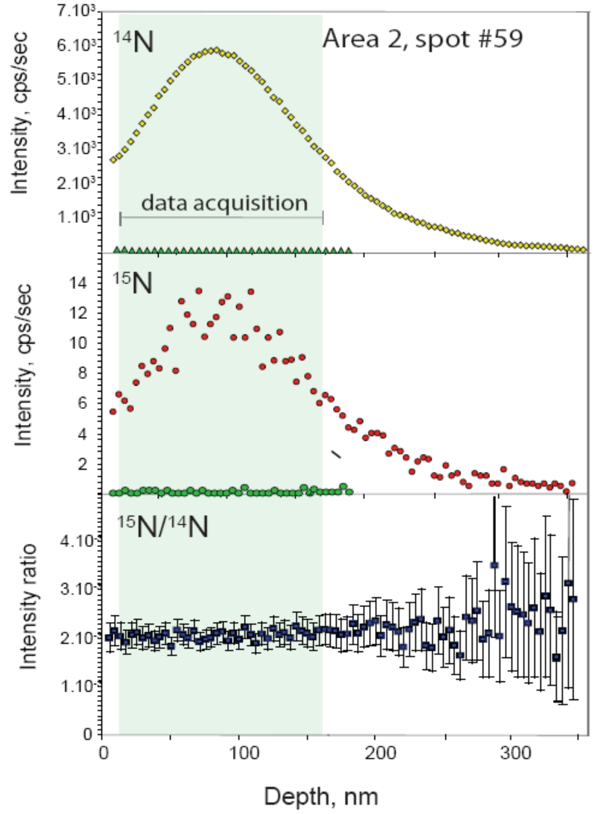

Fig. 1: example of a spot analysis in area 2. N isotopes were analyzed as CN-. Green symbols represent the respective N isotope blanks analyzed on a non flight SiC target material. For N isotope ratio computations, only data within the light green areas were used to obtain an average for each spot. Then for each area an error-weighted average was computed using all spot data for this area.

Our measurements yield a ^{15}N/^{14}N ratio for the SW of 2.178±0.024 $\times 10^{-3}$ (95 % confidence level), corresponding to δ^{15}N = -407±7 ‰ relative to the terrestrial standard (ATM, atmospheric N_2 with ^{15}N/^{14}N = 3.676 $\times 10^{-3}$). By its high precision, this result definitely settles the debate on the N isotopic composition of solar wind in showing that it is extremely ^{15}N-poor compared to other solar system components. The Sun's bulk ^{15}N/^{14}N ratio can be obtained from our Genesis SW measurement once the isotopic fractionations taking place in the convective zone of the Sun and during acceleration of SW are corrected for. We tentatively assign a SW fractionation factor of 1.032 (±0.005) for N isotopes due to Coulomb drag [7], and a 1.007 (±0.002) fractionation factor due to solar diffusive element settling plus radiative levitation [7,8]. Thus, our estimate for the ^{15}N/^{14}N ratio of the bulk Sun, after correction for solar processing and propagation of all errors, is 2.264±0.028 $\times 10^{-3}$ (95 % conf.) (δ^{15}N = -384±8 ‰).

Cosmochemical implications: Several lines of evidence suggest that this solar ^{15}N/^{14}N ratio is representative of the proto-solar nebula (PSN) value. First, this ratio is comparable to that measured in Jupiter's atmosphere (^{15}N/^{14}N = 2.3±0.3 $\times 10^{-3}$ [9]). Second, it is close to the ^{15}N/^{14}N ratio of 2.36±0.04 $\times 10^{-3}$ found in a grain of osbornite (TiN) embedded in a calcium-, aluminium-rich inclusion (CAI) from the CH/CB chondrite Isheyevo [10]. In detail, osbornite appears enriched in ^{15}N by 43±22 ‰ relative to the PSN value. Though presumably negligible isotopic fractionation took place during high temperature condensation, the reservoir in the accretion disk from which the osbornite condensed might have been slightly modified from the PSN composition (this is quite common in the case of oxygen isotopes in CAIs). For instance, this difference could be the result of different mixing proportions in TiN relative to the Sun of originally gaseous N_2 low in ^{15}N with refractory dust rich in ^{15}N. The present result also sheds light on the origin of nanodiamonds in primitive meteorites. They contain pre-solar xenon but their C isotopic composition is clearly solar. Their ^{15}N/^{14}N ratio (2.40±0.03 $\times$ 10^{-3} [11]) comparable to PSN now points also to a solar composition, making it possible that a major fraction of them formed from solar system matter, with only a small fraction, hosting exotic Xe, being inherited from other stellar systems. Finally, nitrogen isotopic variations among solar system objects can be understood as resulting from variable mixing between nebular N low in ^{15}N and a ^{15}N-rich cosmochemical end-member, possibly hosted by organics in primitive meteorites. The origin of the latter is a key question for future studies.

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