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IN SEARCH OF THE SOLAR WIND NITROGEN ISOTOPE COMPOSITION : ANALYSIS OF A GOLD PLATE FROM THE GENESIS SPACECRAFT CONCENTRATOR

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Introduction: Nitrogen displays the largest (after H) isotope variations in the solar system materials (excluding pre-solar grains). The causes of these variations are mostly unknown and discussed in term of mixing between different solar, possibly presolar, reservoirs. Hence knowledge of the N isotopic composition of the Sun is important as this would presumably shed light on the composition of a major reservoir, the protosolar nebula. From the analysis of lunar soils exposed at different periods, an allegedly secular variation of the $^{15}\text{N}/^{14}\text{N}$ ratio, was proposed, from a ^{15}N depletion of ≥ 300 ‰ (relative to terrestrial N, e.g., ATM) in soils exposed ~ 2 Ga ago to ^{15}N enrichments of up to 150 ‰ in ≤ 200 Ma soils (e.g., [1]).

Recently several studies concluded that protosolar nebula N was depleted by ~ 400 ‰, from (i) the combined ion probe analysis of H and N isotopes in lunar soil grains ($\delta^{15}\text{N} \leq 240$ ‰, [2]), (ii) the analysis of the Jupiter atmosphere by either infra-red spectroscopy [3] or *in-situ* by the Galileo probe [4], and (iii) $\delta^{15}\text{N}$ values of -400 ‰ for osbornite (TiN) embedded in a CAI [5].

The Genesis solar wind exposure experiment: The Genesis mission [6] sampled solar wind ions during 27 months in space at the Lagrange point L1, by implantation of SW ions in several targets made of pure material and during different SW regimes. Despite a hard landing of the sample capsule, target material could be recovered, giving new insights into the noble gas composition of the different solar wind regimes [7, 8]. Determining the nitrogen isotopic composition of the solar wind, which is a main scientific goal of the mission, is complicated by the very low amount of implanted solar wind N (fluence of 2×10^{-12} mol/cm² ^{14}N) compared to potential terrestrial contamination. The latter was exacerbated by the capsule crash since nitrogen is a major constituent of the atmosphere, of organics, and of sediments. Decontamination procedures are crucial and those used for samples analyzed for N include washing, UV-ozone cleaning and handling in class 100 rooms. We have developed at CRPG Nancy, France, a ultra high vacuum line and static mass spectrometry, where N is extracted by laser (wl : 193 nm) ablation with a spot size of approx. 50×150 μm rastered over surfaces up to 1 cm². By

modulating either the power of the laser beam, or the number of pulses per area, we are able to remove sequentially layers of matter with a depth resolution of a few nm. The release of SW gases is monitored by the simultaneous analysis of noble gas (He, Ne, Ar) abundances and isotopic ratios. Gold-plated targets were chosen because of the presumed purity of Au layers and of the good extraction efficiency of nitrogen from gold, checked using artificially implanted targets with known ^{15}N fluence.

We analyzed recently gold-over-sapphire (AuoS) target fragments [9] and could not find evidence for a light (^{15}N -depleted), Jupiter-like, nitrogen component. The comparatively large data uncertainties due to low amounts and large corrections for terrestrial contamination did not permit to derive a firm value for the SW N isotopic ratio. We have analyzed further samples, focusing our effort on target material exposed in the Genesis SW ion concentrator and more enriched in SW ions.

Sample and analytical: The Genesis concentrator is a ion-focusing experiment designed to enrich solar wind ions [10] by up to a factor of 30 [11]. It was originally developed for the 3-isotope analysis of SW oxygen in different targets (Fig. 1). The latter were mounted in a gold-plated stainless steel frame, the gold cross. N together with He, Ne and Ar were extracted by laser ablation of one gold cross arm (GCA) (Fig. 1). Five areas of 4-6 mm² were ablated from the border to the center of the GCA. Released gases were split into 2 fractions for nitrogen purification and cryogenic separation/purification of noble gases, respectively. The concentration and isotopic composition of SW neon were analyzed previously at ETH Zürich, Switzerland, along the same arm (laser ablation, spot size of 100 μm), giving us a monitor of SW abundance and isotopic fractionation as a function of location along the arm [11]. Our Ne contents were found to be similar to those determined by [11] within 20 %, despite the large differences in the ablated surfaces. The procedural blank for nitrogen was $1.1 \pm 0.1 \times 10^{-12}$ mol ^{14}N , with $\delta^{15}\text{N} = -22 \pm 15\%$. Together with the flown gold cross arm, we analyzed a spare gold cross arm, which yielded on average $2.64 \pm 0.11 \times 10^{-12}$ mol/mm² with

$\delta^{15}\text{N} = 14 \pm 9\%$. Blanks were negligible for noble gases, which isotopic compositions showed an almost pure SW origin.

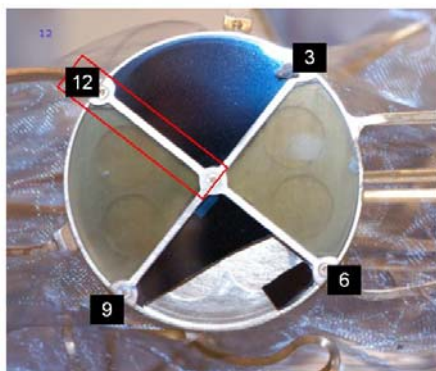


Fig.1: The central target holder ("gold cross") of the Genesis concentrator. The "12 à'clock" arm was analysed.

Results: For evaluating the fraction of SW N in the total N, we use a mixing diagram: $\delta^{15}\text{N} = f[(^{20}\text{Ne}/^{14}\text{N})_{\text{Sample}}/(^{20}\text{Ne}/^{14}\text{N})_{\text{SW}}]$ (SW $^{20}\text{Ne}/^{14}\text{N} = 0.50 \pm 0.09$ from [12]). In this format, mixing between two different end-members yields a unique straight line. Data have been corrected for isotopic fractionation of SW N, from Ne isotope data from [11] and assuming mass dependant functionality for both Ne and N isotope fractionations. Only the fraction of nitrogen that was estimated to be of solar wind origin from the $^{20}\text{Ne}/^{14}\text{N}$ ratio was isotopically corrected. Based on Ne isotopic fractionation, the potential isotopic shift for nitrogen is $\sim 40\%$ at most, which is small compared to for example the 400% difference between terrestrial N and Jovian atmosphere N.

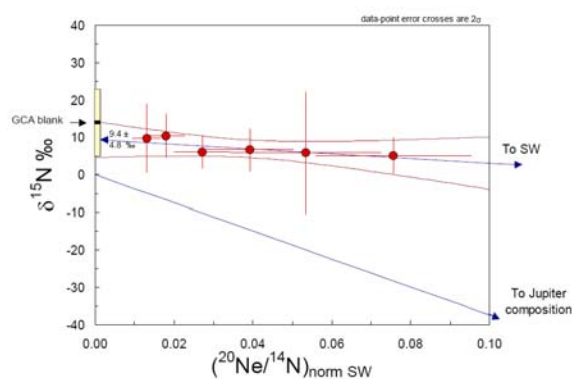


Fig. 2: $\delta^{15}\text{N}$ isotopic composition vs. the Ne/N ratio normalized to the SW value. Dots represent total data for each area along the GCA arm.

Data (either stepwise release ones or those for total release, Fig. 2), define a 2 end-member correlation. One of these has no neon and a $\delta^{15}\text{N}$ value of $+10 \pm 5\%$, which is consistent with the GCA blank value.

Discussion: The other N end-member has an extrapolated $\delta^{15}\text{N}$ value of $-40 \pm 94\%$ (2σ) when all data (previous AuoS targets together with the present results; in Fig. 2 only "total" data for GCA are taken for computing the extrapolated SW value) are considered, which is clearly different from the composition of the Jupiter atmosphere and from the inferred protosolar nebula value. It is, however, in agreement with a $\delta^{15}\text{N}$ of $+40 \pm 8\%$ for recent SW from the analysis of soils exposed in the last few Ma [13]. The present value encompasses also N isotope compositions found in most primitive meteorites (except the CR clan), the Earth, Mars (interior) and Venus.

The difference between modern SW and Jupiter atmosphere is particularly striking because both reservoirs are gigantic and should have averaged local heterogeneities. Thus if the Jovian composition is not representative of the protosolar nebula one, then processes of N isotope fractionation specific to this atmosphere, or having acted during trapping of N in parent icy planetesimals, needs to be explored. Another possibility is a large scale N isotopic heterogeneity in the nascent solar system originating from an N speciation gradient (e.g., [14]).

If the presumed SW N isotope ratio determined in this study is representative of the solar wind composition through time, it is also necessary to find not only one, but two other components trapped in lunar soil, one enriched in ^{15}N which can be accounted by IDPs [2], and the other one depleted in ^{15}N , such as terrestrial atmosphere ions implanted in the past [15].

We are presently working on further concentrator samples cleaned differently with the aim of improving precision on the N isotopic composition of the modern solar wind. Study funded by CNES, CNRS and Région Lorraine.

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