In search of solar wind nitrogen in Genesis material : Further analysis of a gold cross arm of the concentrator.

B. Marty[1], L. Zimmermann[1], P.G. Burnard[1], D.L. Burnett[2], V. S. Heber[3], R. Wieler[3], P. Bochsler[4], R.C. Wiens[5], S. Sestak[6] and I.A. Franchi[6]. [1] CRPG-CNRS, Université de Nancy, BP20, 54501 Vandoeuvre Cedex, France; (bmarty@crpg.cnrs-nancy.fr). [2] California Institute of Technology, Pasadena, USA. [3] ETH Zürich, Switzerland. [4] Universität Bern, Switzerland. [5] Los Alamos National Laboratory, USA. [6] Open University, Milton Keynes,. UK.

Introduction: Determining the isotopic composition of solar wind (SW) nitrogen is one of the top priorities of the Genesis mission. The CRPG (Nancy, France) has developed the analysis of nitrogen and noble gases of gold-plated targets by laser ablation - static mass spectrometry. Last year we presented [1] new N-noble gas data for a few fragments of gold over sapphire (AuoS) target material and for the radial arm of the Genesis concentrator sample holder (gold cross arm, GCA1), a device designed to concentrate the solar wind flux by up to a factor of ~50 [2]. While the AuoS data had large uncertainties, precluding conclusions on the SW N composition, GCA1 analyses showed that the sample contained a large contribution of terrestrial N, as for AuoS targets. However, it also suggested a negative SW N end-member, but within the range of values found in inner solar system reservoirs. Since then, we have analysed another gold cross arm (GCA2) with improved precision.

Experimental: The analytical protocol consisted in rastering surfaces of variable size using a UV laser beam with adjustable energy and thereby excavating the gold substrate to release SW atoms. Tests using blank samples and ^{15}N implants showed that nitrogen is released as N_2 together with noble gases. Aliquots were purified separately, using a CuO furnace and a cold trap in the case of nitrogen, and by classical gettering for noble gases. All gases were analysed sequentially with a modified noble gas static mass spectrometer. Molecular nitrogen was analysed at masses 28, 29 and 30, and the relative abundances of ions at masses 29 and 30 were used to evaluate the purification of N_2 from CO isobars (hydrocarbon masses could be separated thanks to the good mass resolution of the mass spectrometer). In the case of AuoS samples, surfaces up to 1 cm^2 were rastered in order to obtain enough SW N for analyses. However, purification of the released gas was difficult to achieve completely and corrections for mass interferences resulted in large uncertainties up to several tens of ‰. In the case of the GCAs, the enrichment of SW ions permitted rastering of smaller surface areas of the order of a few mm^2. This allowed perfect purification and marginal correction for interferences. Consequently, uncertainties were reduced to a few ‰ (typically 5-6 ‰ at the 2 σ level for N quantities of ~3 x 10^{-11} mol). Neon and argon were analysed together, and noble gas isotopic ratios were used to monitor the occurrence of SW ions and to correct for mass discrimination. Nitrogen blanks were below 1 x 10^{-12} mol and amounted for at most a few % of the analysed N. Experimental blanks and terrestrial contamination were negligible for noble gases.

The efficiency of nitrogen extraction was checked using targets implanted with ^{15}N at energies comparable to that of SW N. It was found to be close to 100 % when corrected for backscattering loss during implantation.

Results: Several areas of typically 4 mm^2 were rastered along GCA2, from the outer edge of the concentrator where the ion concentration was only a factor of 5 to the concentrator center were the ion concentration was maximal (x50). The concentrator is an electrostatic ion focusing device [2] and therefore induces mass/charge discrimination that needs to be corrected for. The mass discrimination for the Ne isotopes and the Ne abundances along the two GCAs we analysed had been determined previously at ETH Zürich [3]. The Nancy data are consistent with these earlier analyses as they show within uncertainties the same mass discrimination effect from the edge to the center of the concentrator, within uncertainties (Fig. 1).

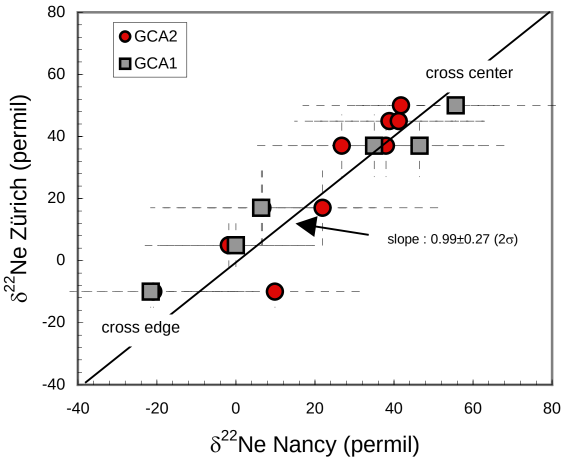

Fig. 1: Comparison of Ne isotope analysis in ETH Zürich (100 μm dots) and in CRPG Nancy (few mm^2 ablation).

Thus we corrected the SW fraction of nitrogen, estimated from the $^{14}N/^{20}Ne$ ratios (see below) for such effect assuming mass dependent fractionation for both Ne and N isotopes. Note that this correction applies only to the few % of SW N within total N, so that the overall correction on bulk $\delta^{15}N$ was small (<2 ‰) and within analytical uncertainties. Such correction does not define, or influence significantly, the correlation depicted in Fig. 2. For the pure SW N end-member, the overallcorrection for concentrator mass discrimination is ~40 ‰ for N isotopes based on Ne isotopic shifts, that is, well within uncertainties on our estimate of the SW N ratio.

In all runs, the amount of extracted N was much in excess of the fluence of SW N during the irradiation time, as observed previously for AuoS and GCA1 runs. In an attempt to improve the SW/contaminant ratio, GCA2 was ozone-cleaned at the Open University using the procedure of [4], a process that proved to be efficient in removing the so-called "brown stain" (a Si-, C-rich layer covering most of target material and attributed to degassing within the spacecraft cell en route to the sampling point). Despite this, all target materials including GCA2 appeared contaminated, presumably by terrestrial N. This contamination is not limited to the surface of gold, but is volume-correlated and within the gold layer itself since our laser ablation steps did not show a significantly decrease of the N/noble gas ratio with depth. Thus correction for the terrestrial N contamination is possible by the use of the SW noble gas/nitrogen ratio, since noble gases are entirely from the SW. In case of a binary mixing between SW N and contaminant N, data will align along a straight line in a $\delta^{15}N$ vs. $^{20}Ne/^{14}N$ space. Thus estimating the SW $^{20}Ne/^{14}N$ ratio is critical in this approach. Last year we used a value of 0.50 from SW $^{20}Ne/^{14}N$, from ref. [5]. However, several lines of evidence suggest that this value might be too low, and not representative of the SW regimes sampled during the Genesis exposure. Here we adopt a $^{20}Ne/^{14}N$ ratio of 1.14, with an uncertainty of 20 %, calculated from the coronal Ne and N abundances derived from solar energetic particles by [6] (with Ne/O = 0.152, N/O = 0.124, as given by [6], and $(^{20}Ne/^{22}Ne)_{SW} = 13.8$). We expect no, or at most a very weak, fractionation between corona and solar wind among the high-FIP elements N, O, and Ne. In fact and despite controversies about absolute photospheric abundances of medium mass elements, the recent N/O photospheric abundance ratios of [7] and [8], and the older value of [9] agree among each other, and are all consistent with our adopted ratio. Within uncertainty, the value of 1.14 is in agreement with in-situ solar wind analyses: a ratio of 1.24±0.30 has been obtained from the ACE mission (D. B. Reisenfeld, pers. comm.), and a value of 0.92 can be estimated as average from Ulysses from the data of four ~300 day periods reported by [10] (two sampling slow and two fast solar wind, respectively).

The new GCA2 data define a negative correlation between $\delta^{15}N$ and $(^{20}Ne/^{14}N)_{norm\ SW}$ (Fig. 2). The $\delta^{15}N$ end-member contaminant (for $^{20}Ne/^{14}N = 0$) is 13 ± 4 ‰ (2σ), consistent with a mean value of 14 ± 9 ‰ analysed for a spare GCA and within the range of terrestrial values. The $\delta^{15}N$ end-member for $(^{20}Ne/^{14}N)_{norm\ SW} = 1$ (pure SW N) is around -400 ± 180 ‰ (2σ), within the range of the Jupiter atmospheric value [11]. This result suggests that SW N is indeed light and did not evolve with time from the protosolar nebula value. It also calls for a large scale N isotope heterogeneity in the Solar system between the protosolar gas on one hand and N compounds in inner planets, meteorites, and comets on another hand.

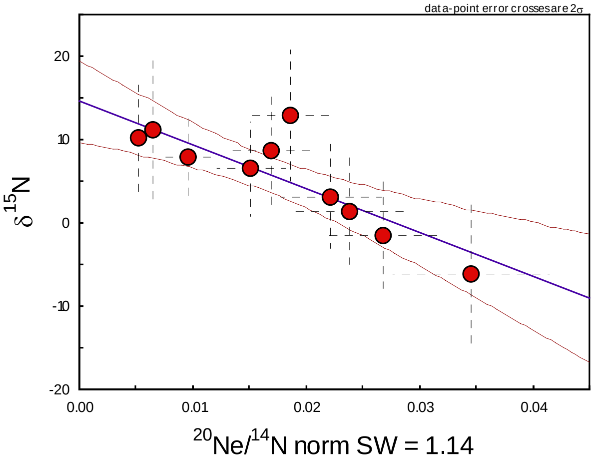

Fig. 2: GCA2 data. Each data point is corrected for mass discrimination using the conjointly measured $^{20}Ne/^{22}Ne$. This correction is < 2 ‰ and cannot account for the correlation. Note that the correlation will yield a negative $\delta^{15}N$ value whatever the adopted value of $(^{20}Ne/^{14}N)_{SW}$ ratio.

[1] Marty B. et al. (2008) *LPS XXXIX*, Abstract #1314. [2] Wiens R.C. et al. (2003) *Space Sci. Rev. 105*, 601. [3] Heber V.S. et al. (2007) *Space Sci. Rev. 130*, 309-316. [4] Sestak S. et al. *LPS* XXXVII, Abstract #1878. [5] Gloeckler G. and Geiss J. (2007) *Space Sci. Rev. 130*, 139. [6] Reames D. V. (1995) *Adv. Space Res. 15*, (7)41. [7] Asplund M. et al. (2006) *Nucl. Phys. A 777*, 1. [8] *Lodders K. (2003) Ap J. 591*, 1220. [9] Anders E. and Grevesse N. (1989) *GCA. 53*, 197. [10] Von Steiger et al.(1995) *Adv. Space Res. 15*, 3 [11] Owen T. et al. (2001) *ApJ 553*, L77.