CARBON, NITROGEN, AND OXYGEN ABUNDANCES IN THE BULK SOLAR WIND AND CALIBRATION OF ABSOLUTE ABUNDANCES. V.S. Heber¹, D.S. Burnett², J. Duprat³, Y. Guan², A.J.G. Jurewicz⁴, B. Marty⁵, K.D. McKeegan¹: 1 Dept. Earth and Space Sciences, UCLA, Los Angeles, CA, USA, heber@ess.ucla.edu; 2 CalTech, Pasadena, CA, USA; 3 CSNSM IN2P3-CNRS, Orsay Campus, France; 4 Arizona State University, Tempe, AZ, USA; 5 CRPG-CNRS, Nancy, France.

Introduction: Carbon, nitrogen and oxygen abundances measured in the solar wind (SW) are an important constraint for the abundance of these elements in the Sun. Photospheric spectroscopy measurements have large uncertainties and C, N, and O abundances have been revised downward over the last two decades [1, 2]. The thus reduced solar metallicity lead to a still-unresolved disagreement with solar models based on helioseismology [see 3]. Spacecraft measurements of SW yield C, N, and O abundance ratios with uncertainties of 10-50% [e.g., 4, 5]. The objective of the Genesis mission is to provide abundances of these elements with an accuracy of 5% and better.

Solar wind fluences that are measured by secondary ion mass spectrometry (SIMS) are generally calibrated against commercial implants of reference ions [6-9]. However, ion implant facilities have difficulty quantifying the errors in the fluence of the implanted ions; an uncertainty of 10% is generally assumed. Here, we present the most recent results on C, N and O fluences in the SW, as well as our strategies to determine absolute fluences using calibrated implants.

CNO abundances in the bulk SW: We analyzed C, N and O fluences in Genesis Si bulk SW collectors by back-side depth profiling using SIMS in two sessions in 2010 and 2012. Analytical conditions are described in [6]. In total we analyzed 3 backside polished Genesis targets thinned to 400nm, 2.5µm, and 1µm, respectively. The latter was the optimal thickness for analysis. Additionally, we performed rigorous interference tests for N (measured as ²⁸Si¹⁴N⁻) which were still pending in [6]. The fluences (Table 1) were calibrated with reference implants: for ¹⁶O we used an ¹⁸O implant by Leonard Kroko Inc. (LK 09/2007), for ^{14}N a ^{15}N implant (LK 09/2007), and for ^{12}C a ^{13}C implant by Los Alamos Nat. Lab. (LANL 04/2005). Mass fractionations between the isotopes were not considered in the final fluences as they are smaller than our given errors.

Table 1. Bulk Solar Wind C, N, O fluences

	Fluence and standard	number of
	error (atoms/cm ²)	analyses
¹² C	$(6.35 \pm 0.17) \times 10^{12}$	9
¹⁴ N	$(1.14 \pm 0.06) \times 10^{12}$	7
¹⁶ O	$(1.20 \pm 0.06) \times 10^{13}$	5

The inferred SW N fluence disagrees with the value given by [7], who used material from the same ¹⁵N ion implant as standard but obtained twice as high SW N fluence. In the latter work, Genesis targets were conventionally analysed by sputtering the frontside with a 20keV primary Cs⁺ beam. Under these conditions the SW profile is entirely masked by surface contamination gardened-in by the primary beam and fluences were estimated using a model for implanted SW ions (Fig 1a). In contrast, the signal obtained from the backside depth profiling with a low energy primary ion beam [6]) results in a nearly complete and contamination-free SW profile (Fig. 1b)

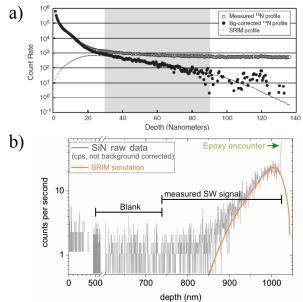


Fig. 1. ¹⁴N profiles measured in bulk SW by a) sputtering the target from frontside using a 20keV primary Cs^+ beam [7] and b) sputtering from the backside with a 5keV primary Cs^+ beam (this work). The latter represents an almost complete SW profile not affected by the gardening-in of surface contamination.

Cross-calibration of the reference ion implants: To determine the absolute fluences of the ion implants used as standards, we first attempted to cross calibrate four reference ion implants from different ionimplantation facilities. Each implant was measured in the center of a 7mm-center-hole holder. Samples were exchanged after each measurement. The reproducibility of the integrated signal (fluence) of a given implant was between 0.5 and 2%. The implant we used to calibrate the Genesis fluences was designated as the "anchor" (shown in gray in Table 2) to which the other ion implants were related.

Table 2. Percent deviation of measured fluences of ion implants from the nominal fluence of the "anchor"ion implant

¹⁸ O		¹³ C		¹⁵ N			
% deviation from							
LK 09/2007		LANL 04/2005		LK 09/2007			
LK		LK		Cutting			
08/2008	7 ± 2	08/2008	18 ± 2	Edge Ions	-16 ± 1		
Hughes		Cutting		Hughes			
Res. Lab.		Edge		Res. Lab.			
09/1999	-22 ± 2	Ions	0 ± 1	09/1999	-36 ± 1		
CORE		CORE		LK			
Systems	-10 ± 3	Systems	-5 ± 1	11/2012	-12 ± 1		

The fluences measured for the four ion implants for each element show a spread from nominal values by ~30% for ¹⁸O, 23% for ¹³C, and 36% for ¹⁵N. For unknown reasons, carbon shows the best reproducibility with 3 out of 4 ¹³C implants from different implant facilities agreeing within 5%. Thus, it is tempting to assume that these nominal fluences are indeed the correct values, but we cannot be sure. Therefore, these results are not yet included in the bulk SW C, N, and O fluences given in Table 1.

Additionally, the knowledge that a nominal fluence asked for and implanted by ion implantation facilities can vary as much 36% is important to consider when SW fluences for other elements are given that can be measured with high precision, as e.g. Al [9]. What are the reasons that implanted fluences deviate from their nominal values? Three obvious factors could play a role: i) the measurement of the current of the ion beam hitting the target, ii) the purity of the source material used for the ions, and iii) unaccounted interferences due to a poor vacuum ($\sim 10^{-6}$ torr) and mass resolution of (0.5 mass units) in the ion implanter. According to Leonard Kroko Inc. the current measurement is calibrated regularly at their facility.

In summary, the ion-implant cross-calibration experiment showed that a large uncertainty potentially accompanies any "standardization" using the nominal value given for the fluences of a reference ion implant not otherwise certified (e.g. NIST). Other measures must be employed to provide a precise, absolute calibration of the SW fluences. Non-volatile elements that occur as trace elements in terrestrial materials can be used for quantitation of implanted fluences following the method of [10], however this method cannot be easily applied to calibrate the fluences of C, N and O. These elements rarely exist in homogeneously distributed trace quantities in terrestrial materials.

Calibration of a nitrogen ion implant: For N we will employ gas mass spectrometry to calibrate the fluence of a ¹⁵N ion implant (LK 11/2012). ¹⁵N was implanted in two gold targets that were surrounded by Si targets. The nominal ¹⁵N fluence is 1×10^{15} ions/cm². Ions were implanted with an energy of 100keV. Using SRIM [Stopping and Range of Ions in Matter, 11] we calculated a backscatter loss of ¹⁵N from Au of 16%. We demonstrated in [12] for Ne implantation into Au that backscatter loss under these conditions can be precisely modeled with SRIM. The Au targets will be analysed by total extraction in an oven using a gas mass spectrometer and the fluence inferred from precise area measurement of the Au target. Analysis of the implant in Si targets by SIMS showed that no ¹⁴N is implanted with ¹⁵N. The ¹⁵N background can thus be determined and subtracted via ¹⁴N using the terrestrial N isotopic composition. Measurements will be carried out at CRPG CNRS in France by B. Marty.

Carbon and Oxygen calibration. To calibrate O and C fluences we employ an ion implantation performed at CSNSM using SIDONIE, an electromagnetic mass separator built for production of high purity targets for nuclear physic experiments [13]. C and O implantation are performed with monocharged ions produced at 40keV by a hot cathode ion source (using ¹³C and ¹⁸O enriched gases) and analyzed in a large radius magnet. The beam intensity is monitored with a precision of a few % by faraday cups and induced current measurement at the focal plane and at the target position. Potential interfering isotopes and molecules will be quantified using mass scans performed before and after the implantation process. With all possible sources of uncertainties tracked and quantified, we should obtain implantation with accurate nominal fluences

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References: [1] Anders, E. and N. Grevesse (1989) Geochim. Cosmochim. Acta, 53: p. 197-214. [2] Asplund, M., et al. (2009) Annu. Rev. Astron. Astrophys., 47: p. 481-522. [3] Bochsler, P., et al. (2013) in 44th LPSC. p. 1277. [4] von Steiger, R., et al. (2000) J. Geophys. Res., 105: p. 27217-27238. [5] von Steiger, R., et al. (2010) Geophys. Res. Letters, 37: p. L22101. [6] Heber, V.S., et al. (2011) 42nd LPSC, The Woodlands, Tx, USA, #2642. [7] Huss, G.R., et al. (2012) Meteoritics & Planetary Science, 47: p. 1436-1448. [8] Veryovkin, I.V., et al. (2012) in 43rd LPSC. p. #2296. [9] Heber, V.S., et al. (2013) in 44th LPSC. [10] Leta, D.P. and G.H. Morrison (1980) Analytical Chemistry, 52: p. 277-280. [11] Ziegler, J.F. (2004) Nuclear Instruments and Methods in Physics Research, 219/220: p. 1027-1036. [12] Heber, V.S., et al. (2011) Meteoritics & Planet. Sci., 46: p. 439-512. [13] Chauvin, N., et al. (2004) Nucl. Instr. & Meth. in Phys. Res. Sect. a -Accelerators Spectrometers Detectors and Associated Equipment, 521: p. 149-155.