OXYGEN ISOTOPES IN A GENESIS CONCENTRATOR SAMPLE. K.D. McKeegan[1], A.P. Kallio[1], V. Heber[1], G. Jarzebinski[1], P.H. Mao[2], C.D. Coath[3], T. Kunihiro[4], R. Wiens[5], J. Allton[6], and D.S. Burnett[2] [1] Dept. of Earth & Space Sciences, UCLA, Los Angeles, CA. 90095-1567 USA, [2] Div. Geol. & Planetary Sci., Caltech, Pasadena, CA 91125, USA. [3] Dept. of Earth Sciences, Univ. of Bristol, Bristol, BS8 1RJ, UK, [4]SEI, Okayama Univ., Misasa, Tottori 682-0193 Japan, [5] Los Alamos National Laboratory, Los Alamos, NM 87545 USA, [6]Johnson Space Center, Houston, TX 77058 USA.

Introduction: Oxygen is the major constituent of rocky planets and the third most abundant element comprising the Sun, yet the solar oxygen isotopic composition has remained essentially unknown. One reason is that the usual appeal to primitive meteorites does not work because oxygen is isotopically distinct in all different classes of meteorites [1]. The cause of this premier "isotopic anomaly" has been variously ascribed to nucleosynthetic input, e.g. from a nearby supernova, or to exotic isotope-selective chemistry inthe solar nebula, e.g. based on molecular symmetry or UV photolysis [2-7]. Knowledge of the average starting composition of the solar system, which is preserved in the Sun, would provide a baseline from which one could interpret the oxygen isotopic compositions of planetary materials. To this end, NASA flew the Genesis Mission to capture samples of the solar wind (SW) in ultra-pure target materials and return them to Earth for laboratory analysis [8]. At UCLA, we have designed and constructed a hybrid secondary ion and accelerator mass spectrometer (SIMS/AMS), called the "MegaSIMS", specifically to tackle the unique analytical challenges posed by the Genesis samples: dilute elemental concentrations, limited sample material, and close proximity of likely surface contamination to the implanted SW ions [9]. We reported our analytical protocol and preliminary data at the LPSC last year [10]; here we provide additional details and we plan to report the results of further measurements to characterize the isotopic composition as a function of radial position on the concentrator collector at the meeting.

Genesis Concentrator sample #60001: Eight analyses were performed on Genesis concentrator SiC sample #60001. Prior to insertion in the custom-made quadrant sample holder of the UCLA MegaSIMS, the sample was ultrasonically agitated in xylene which removed most of the coarse particles adhering to the surface. Optical inspection of the sample confirmed that some areas were damaged and/or contaminated in the crash, but that relatively large areas are free of particles larger than ~1μm. So long as their number density is small, particles in this size range remaining after ultrasonic cleaning can be rapidly recognized by ion microscope imaging at the beginning of a depth profile analysis. The sample can then be shifted such that these contaminants are outside the analytical area,

which is gated to a ~100 μm diameter imaged field by an aperture inserted in the first image plane at the entrance of the (low-energy) mass spectrometer.

After insertion in the MegaSIMS vacuum, sample #60001 was left in the sample chamber for 5 days during which time the chamber was baked at 125 ºC for 46 hours. The immersion lens extraction plate was cleaned prior to each day's analysis by intense (~80nA) Cs-sputtering of blank SiC for 4 hours. For the initial measurements, we picked a location close to a defect in the SiC, so as not to consume area that would be valuable for other analyses (r = 20mm, θ = 45º). A second series of analyses were made approximately 9mm closer to the origin, where O concentrations were a factor of ~3 higher.

Prior to each analysis, the sample surface in a ~200 μm diameter circle surrounding the targeted area was cleaned by Cs sputtering with 5 keV impact energy at 20 nA for 5 minutes. This procedure removes ~20 nm of the surface [9,10], but it very effectively reduces the impact gardening from an e-folding length of ~7.5 to ~1.0 nm, thus removing residual oxygen contamination from the first few atomic layers of the sample without appreciably mixing it into the implanted SW. Analyses were carried out with a Cs^+ beam rastered over 130×130 μm^2 at intensities ranging from $20 - 30$ nA depending on the oxygen concentration (which varies with radius on the collector).

A typical O^{++} profile is shown in Figure 1. Sputtering continued through the implanted solar wind layer (mean depth ~ 75 nm) until a steady-state background due to residual oxygen from the vacuum ($<3\times10^{-11}$ Torr) was reached (typically depths > 250 nm). Under these conditions, the integrated background is <2% of the solar wind signal. Background-corrected solar wind depth-profiles are well-fit by a Gaussian distribution as expected due to the multiple charge states and impact angles onto the concentrator target. Also expected is the small difference in mean implantation depth which increases by ~1.5 nm with increasing isotopic mass and width of the distribution which also increases slightly at higher mass due to higher implantation energy. Fit parameters are used to normalize integration areas for each isotope and data are corrected for deadtime, detector efficiency, and instrumental mass fractionation using a terrestrial standard.
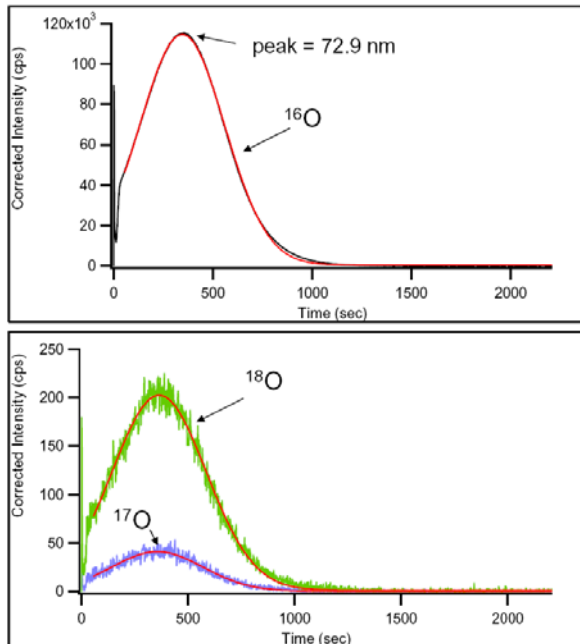
Fig. 1. Oxygen depth-profiles from Genesis SiC sample 60001 plotting intensity of O^{++} signals as a function of sputter time. Red line shows Gaussian fit to data. Time is converted to depth by known sputter rate. The first ~21 nm are missing due to cleaning.

Results: All analyses indicate an ^{16}O-rich composition for the captured solar wind with a mean $\Delta^{17}O = -26.5 \pm 5.6$ ‰ (2σ). The two areas of the collector are mass fractionated relative to each other by a large amount, ~20‰ /amu. This is within the range expected based on models of the concentrator optics [11] and is consistent with Ne isotopic measurements made as a function of radial distance in the Au-cross retaining bar [12]. A correlated radial traverse is planned for oxygen and neon isotopes which will enable a robust correction for isotopic fractionation induced by the electrostatic concentrator by comparison of $^{22}Ne/^{20}Ne$ measured in SiC sample #60001 with that measured in a bulk (diamond-like carbon on Si) solar wind target. Until this correction is quantified, the agreement of most of the preliminary data presented here with the CAI-mixing line (Fig. 2) must be considered fortuitous. Other sources of mass-dependent fractionation, e.g., in the acceleration of the solar wind and, possibly, in gravitational settling in outer layers of the Sun, will also induce shifts in extrapolating the solar wind composition to solar values. However, these corrections are expected to be mass-dependent and thus will not change the first-order conclusion that the Sun is ^{16}O-enriched relative to Earth and other planetary materials.
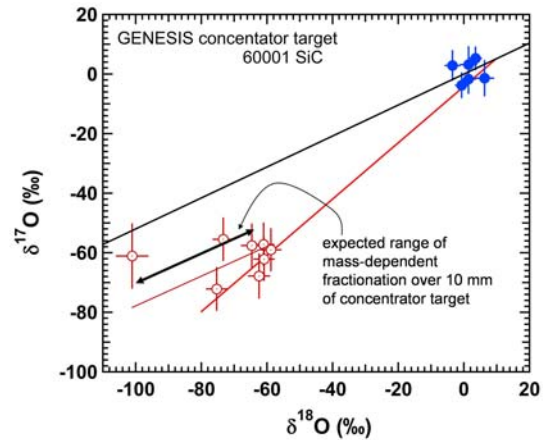

Fig. 2. Preliminary data for oxygen isotopic composition of solar wind on Genesis SiC sample 60001. Red points indicate integral intensities for ~100 μm diameter areas analyzed; blue points are terrestrial standards. Uncertainty estimates are based on counting statistics. The approximate expected range of mass-dependent fractionation induced by the electrostatic concentrator is indicated by the arrow. Overall mass fractionation remains to be calibrated.

Implications: The data are in semi-quantitative agreement with expectations based on models of chemical and isotopic evolution of oxygen in the solar nebula or its precursor cloud by isotope-selective photochemical self-shielding of CO [5-7]. Other models invoking chemical non-mass-dependent isotope fractionaton are not ruled out [3-4], however those considering nucleosynthetic input from nearby massive stars are problematic [see 13]. The maximum $\Delta^{17}O$ seen for Genesis is close to that measured by Kobayashi et al. [14], which is more ^{16}O-enriched than most CAIs.

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