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SOLAR AND SOLAR-WIND OXYGEN ISOTOPIES AND THE GENESIS MISSION  
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Introduction: The oxygen isotope heterogeneity exhibited by a wide variety of solar system materials is not understood. Various models that will likely be discussed in this oxygen isotope session include 1) partial equilibration of $^{18}$O-enriched solids with isotopically heavier gases in the solar nebula to produce the slope-1 mixing line on the oxygen three-isotope plot [1,2]; 2) non-mass-dependent fractionation by gas-phase chemical reactions to produce a trend with a slope of exactly 1.0 [e.g., 3]; 3) oxygen isotopic self-shielding [3,4,5] to enrich starting materials in $^{17,18}$O.

It is widely accepted that an accurate oxygen isotope composition measurement for the Sun, which represents >99% of all material in the present-day solar system, would distinguish between these theories, thereby providing important information on the processing of pre-planetary materials in the solar nebula. In the case of the nebular mixing model, the predicted solar composition is heavier than oxygen in meteoritic or planetary bodies [6,7]. Non-mass dependent chemical fractionation predicts a solar composition that is close to the average of meteoritic and planetary materials, i.e., close to the terrestrial fractionation line. On the other hand, isotopic self-shielding would be expected to enrich planetary materials in heavy isotopes of oxygen, in which case the solar oxygen composition would be isotopically light, similar to oxygen in calcium-aluminum inclusions [5].

Measurements to Date: There are two methods of measuring the solar isotopic composition: either by observations of photospheric emission lines from molecular species in low-temperature regions above sunspots, or by measuring the composition of the solar wind. In neither case has the $^{15}$O composition been measured to better than a factor of 2.5. Photospheric emission line observations yielded $\delta^{15}$O = 130 (+145, -115)% (1-σ) [8]. Solar-wind observations from spacecraft instruments have given $\delta^{18}$O = 110 (+450, -250)% (1-σ) for slow wind [9] and 120 (+280, -190)% (2-σ) for coronal hole (fast) wind [10]. These values are shown graphically in Fig. 1. It is clear that neither type of measurement—photospheric emission lines of in-situ measurement of solar wind by spacecraft instruments—will be accurate enough to provide meaningful constraints on the theories attempting to account for solar-system oxygen isotopic heterogeneities. Because of this, the Genesis mission has as its highest priority the measurement of oxygen isotope ratios in collected solar wind, with a goal of ±1 permil precision [11].

We will discuss two significant challenges in attaining this goal: i) the prospects for obtaining a fractionated-corrected value for oxygen isotopes in the solar nebula via the solar wind, and ii) the capability to measure oxygen isotope ratios in the diffusely-implanted solar-wind atoms to the required precision, once the samples are returned to Earth.

Isotopic Fractionation in the Sun and Solar-Wind: Over the past decade the case for gravitational fractionation of isotopes within the sun has become significantly stronger, as models of the interior of the Sun match the seismic data substantially better with a moderate amount of gravitational separation [e.g., 12]. The major factor in the models is the helium abundance, but secular gravitational settling of helium implies isotopic fractionation in general. Current models predict ~33‰ fractionation for $^3$He/$^4$He and -5‰ per amu for oxygen isotopes [13].

Another source of fractionation is the ionization and acceleration of the solar wind. Elemental fractionation of the solar wind relative to the photosphere is both significant and rather well-investigated. Elemental fractionation differs depending on the type of solar-wind, depending on the conditions of ionization and acceleration within the inner corona. Elemental fractionation is based on the first ionization potential (FIP), and varies between factors of ~2 for coronal hole material and ~4 for interstream wind [14]. Fractionation based on ionization characteristics does not affect isotopic ratios, but theories on the acceleration of solar-wind do predict isotopic fractionation. The most widely accepted models for acceleration of minor species involves Coulomb drag. These models attempt to describe the opposing forces of solar gravitation and collisions with protons [e.g., 15]. According to these models, fractionation relative to H of a species of mass number A and atomic charge q should scale as $(2A - q - 1)*[(1 + A)/A]^{1/2} * q^{-2}$ [e.g., 13]. Based on the observed fractionation of He/H, oxygen isotopic fractionation in the interstream wind most common in the ecliptic plane is predicted to be around -25‰ per amu. Coronal hole oxygen fractionation is predicted to be around -8‰ per amu [13]. The evidence for Coulomb drag fractionation, in the form of solar-wind helium isotope fractionation, is quite ambiguous at this time. Significant effort has been made with the Genesis mission to collect separate samples of interstream and coronal hole wind specifically to compare the isotopic ratios from different types of solar wind, and thus understand any isotopic fractionation inherent in the acceleration of the solar wind.
Analysis Plans for Genesis Samples: The Genesis capsule is due to re-enter the Earth’s atmosphere in September, 2004, with samples of solar wind collected over a period of about 27 months [16]. The solar wind is implanted in the top ~100 nm of ultrapure collectors [17]. Oxygen analyses will utilize substrates which exclude oxygen as a trace element, including CVD diamond, diamond-like thin films, and single crystal silicon carbide. Additionally, a special type of silicon wafer, float-zone Si can be used, which is grown under very reducing conditions. However, FZ Si will still tend to develop a surface layer of oxygen under room temperature conditions.

The oxygen abundance averaged over the top 100 nm after 27 months of solar-wind exposure is expected to be ~1 x 10^19 cm^-3 (total fluence of ~1 x 10^13 cm^-2). Analysis of this oxygen to a 1 permil uncertainty will be very challenging. To provide an alternative, a solar-wind concentrator was developed with the goal of increasing the oxygen fluence to a small (~28 cm^2) target by an average factor of 20 [18]. The implanted ions will be fractionated at the several permil level which will vary as a function of target radius [19] but this can be accurately calibrated and verified by precise measurement of the neon isotope ratios, also as a function of the radius from the center of the concentrator target.

The Genesis project has supported several different approaches for oxygen isotopic analyses, including ion probe (UCLA), laser fluorination and mass spectrometric analysis as O2 (UCSD), and mass spectrometric analysis as CO (Open U.). At UCSD the laser fluorination system has been tested, and has been recently shown to give quantitative yields for SiC target materials with isotopically labeled oxygen implants at the 3 x 10^15 cm^-2 fluence level.

In the approach developed by the Open University, isotopic analysis is done on CO produced from oxygen extracted by laser ablation from CVD diamond. To minimize isobaric interference, 13C diamond targets are being flown in the concentrator. Surface cleaning and sample extraction will be performed by UV (193 nm) laser ablation. Preliminary results indicate that suitable depth resolution can be achieved. [23]. System blanks are currently ~20 pg of O2 for a 10 minute extraction (~0.5% of anticipated sample yield). The CO liberated will be purified of condensable gases and analyzed on a static vacuum mass spectrometer. The best analytical precision so far achieved on aliquots equivalent to the anticipated sample size with the existing instrument is ±4% (1σ) for δ18O. However, a major re-development of the ion source to use a cold electron source is expected to improve this by an order of magnitude.

Secondary ion mass spectrometry (SIMS) analyses will be carried out on a newly-developed accelerator-based ion microprobe at UCLA, termed the MegaSIMS. High resolution depth profiles will be made on ~40000 μm^2 areas of concentrator target by Cs bombardment to resolve surficial contaminant oxygen and to extract negative secondary ions from implanted solar wind. A tandem accelerator will be used to destroy hydride and other molecular ions prior to ion counting analysis by multiple collector mass spectrometry.

References:

![Figure 1. δ18O values for the photosphere [8], two types of solar wind [8,9], meteoritic materials, and comet Halley [20,21]. The terrestrial standard (SMOW) lies by definition at 0. δ18O values are not available for the data points shown here. One- and two-sigma uncertainties are as given in the text. After [22].](image)