NUCLEOSYNTHETIC Os ISOTOPIC ANOMALIES IN CARBONACEOUS CHONDRITES. T. Yokoyama[1], V. K. Rai[2], C. M. O'D. Alexander[3], R. S. Lewis[4], R. W. Carlson[3], S. B. Shirey[3], M. H. Thiemens[2], and R. J. Walker[1].

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Introduction: Diverse contributions from various nucleosynthetic sources have been documented in the isotopic compositions of a number of elements in presolar grains residing in bulk chondrites [1]. Presolar grains survived the dynamic evolution of the protosolar accretionary disk via early incorporation into chondritic parent bodies that experienced little subsequent metamorphic processing. The extent and efficiency with which these grains were mixed in the solar nebula are controversial [2-8]. Recently, significant deficits in Os isotopes with large contributions from s-process nucleosynthesis (^{186}Os, ^{188}Os and ^{190}Os) were found in some carbonaceous and possibly enstatite chondrites, when compared to the average of ordinary chondrites and terrestrial Os [9]. Because presolar SiC grains are extremely acid resistant and strongly enriched in s-process nuclides [1], it was concluded that the s-process deficits reflected incomplete acid dissolution of the chondrites with acid insoluble SiC hosting the complementary enriched Os. However, that study did not directly detect the s-process-enriched component, nor could it eliminate the possibility that the s-process deficiencies reflect large-scale Os isotopic heterogeneity in the early solar system.

Experimental: We have precisely measure Os isotopes in bulk samples of carbonaceous (Renazzo, EET92042, Tagish Lake, Murchison and Allende), enstatite (Yilmia and Pillistfer) and ordinary (Dhajala and Allegan) chondrites. These samples were processed using an alkaline fusion total digestion technique (AF) which decomposes diamond and SiC, as well as major mineral phases. We also digested five bulk carbonaceous chondrites with *aqua regia* by applying the conventional Carius tube technique (CT). In addition to bulk chondrites, we examined acid residues of Tagish Lake, Murchison and Allende as well as two nanodiamond fractions from Allende. To access the Os in these fractions, we applied a newly developed combustion technique (COMB) which combusts diamond and SiC at 1000 °C with extremely low blanks. In some cases, aliquots of acid residues were combusted at up to 800°C. The objective of precombustion was to further concentrate Os in the highly-refractory SiC present in the acid residues. Os isotopes were measured by a TIMS (*ThermoElectron Triton*) at the Carnegie Institution of Washington with negative ionization mode.

Results and Discussion: In order to resolve possible nucleosynthetic isotope anomalies, data for all samples were normalized to ^{189}Os and corrected for mass fractionation using ^{192}Os/^{189}Os = 2.527411. The ^{186}Os/^{189}Os^{i} ratios are calculated initial ^{186}Os/^{189}Os ratios that are time-corrected for ^{190}Pt decay over 4.56 Ga. The Os isotopic compositions of all bulk chondrites processed by AF are indistinguishable from each other and terrestrial. We conclude that the average of the measurements define the relative solar abundances of these Os isotopes. For bulk samples processed by CT and acid-resistant residues, we report deviations from these presumed solar values in εOs units (**Fig. 1**). Five bulk carbonaceous chondrites processed by CT, with the exception of Allende, have negative anomalies in ε^{186}Os^{i}, ε^{188}Os and ε^{190}Os that are well resolved from the solar average. The depletions overlap with the range previously reported for Tagish Lake [9]. In contrast, εOs values for acid residues of Tagish Lake and Murchison are strongly positive. Precombusted acid residues from both Allende and Murchison are also enriched, as are two nanodiamond-enriched fractions processed by COMB. Three acid residues of Allende, however, are not resolvable from solar.

The measured isotopic heterogeneities must be attributable to variable contributions from at least two nucleosynthetic components present in the meteorites. In **Fig. 2**, the data define positive linear correlations that pass through the solar values which most likely represent mixing trends between s-process enriched and depleted components. For Murchison and Tagish Lake, the isotopic compositions of the acid residues and the bulk rocks decomposed by CT are complementary. The s-process carrier is concentrated in the acid residues, and it evidently remained undissolved when the bulk chondrites were decomposed by CT. It remains unknown if melted portions of the meteorites, e.g. chondrules and CAIs, have solar Os isotopic compositions or are depleted. If solar, the s-process-depleted, r-process-enriched component must reside in the matrix of these meteorites.

Several factors suggest the s-process carrier is SiC. Direct isotopic analyses of Si and C in single presolar SiC grains have revealed that >90% of SiC grains are 'mainstream grains' that have isotopic signatures dominated by s-process nucleosynthesis [1]. These grains presumably originated from AGB stars. Only a small fraction of SiC grains have isotopic signatures of different origin, such as type II supernovae. Because concentrations of cosmochemically similar elements in presolar SiC grains are extremely high (e.g.

~2 to 57 ppm for Mo and ~1 to 54 ppm for Ru) [10], it is likely that excesses in *s*-process Os isotopes present in the acid residues came from the mainstream SiC grains.

All bulk chondrites decomposed by *AF* have uniform, terrestrial Os isotopic compositions, although some of these chondrites contain isotopically diverse presolar materials. The micro-scale heterogeneity but macro-scale homogeneity indicates that presolar components with heterogeneous Os isotopic compositions were well mixed in the solar nebula, prior to the onset of planetesimal accretion. The extent, timing and processes involved in nebular homogenization are debated. Osmium homogenization could either have occurred via mixing in a turbulent presolar molecular cloud, or by very early large-scale mixing and transport of matter in a marginally gravitationally unstable solar nebula [11]. Analysis of chondrules and CAIs may help to resolve this issue.

Isotopically homogeneous Os in bulk chondrites is consistent with some previous studies that reported uniform isotopic compositions of Zr, Mo and Ru in bulk chondrites[6-8], but may conflict with reports of *s*-process isotope heterogeneity in Mo and Ba in bulk samples of some chondrites[3-5]. Purported isotopic heterogeneities of other elements must either reflect variable contributions from distinct, Os-poor but *s*-process-rich components, or are the result of analytical artifacts that may include incomplete digestion of presolar components. This study, coupled with [9], highlights the serious problem of incomplete sample digestion when processing bulk samples of chondrites. None of the studies reporting isotopic anomalies in bulk chondrites utilized digestion techniques that would ensure the decomposition of acid-resistant presolar components, although some anomalous compositions have been reported for chondrites of higher metamorphic grade that presumably no longer contain intact presolar grains [4]. Further study of these elements using total digestion processes will be needed to resolve this issue.

References: [1] L.R. Nittler. (2003) *EPSL*, *209*, 259-273. [2] R. Andreasen, M. Sharma. (2006) *Science*, *314*, 806-809. [3] N. Dauphas, et al. (2002) *Ap. J.*, *569*, L139-L142. [4] M.C. Ranen, S.B. Jacobsen. (2006) *Science*, *314*, 809-812. [5] Q.Z. Yin, et al. (2002) *Nature*, *415*, 881-883. [6] H. Becker, R.J. Walker. (2003) *Nature*, *425*, 152-155. [7] H. Becker, R.J. Walker. (2003) *Chem. Geol.*, *196*, 43-56. [8] M. Schonbachler, et al. (2003) *EPSL*, *216*, 467-481. [9] A.D. Brandon, et al. (2005) *Science*, *309*, 1233-1236. [10] Z. Kashiv, et al. (2001) *LPS*, *32* 2192. [11] A.P. Boss. (2006) *Meteor. Planet. Sci.*, *41*, 1695-1703. [12] C. Arlandini, et al. (1999) *Ap. J.*, *525*, 886-900.
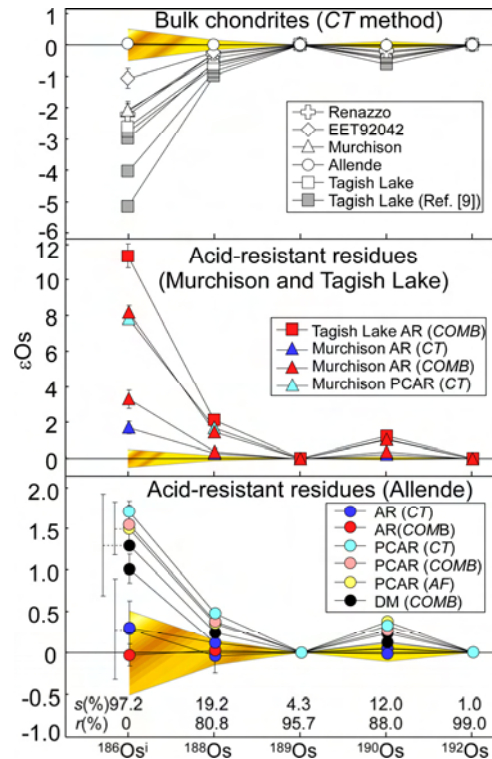
Fig. 1. εOs values for bulk chondrites and acid residues. $\varepsilon^x Os = \{(^x Os/^{189}Os)_{sample}/(^x Os/^{189}Os)_{solar} - 1\} \times 10^4$. Errors are $\pm 2\sigma_m$. Yellow zones are the $\pm 2\sigma$ reproducibilities of nine bulk carbonaceous chondrites processed by AF. Contributions of s- and r-processes are calculated by [12].
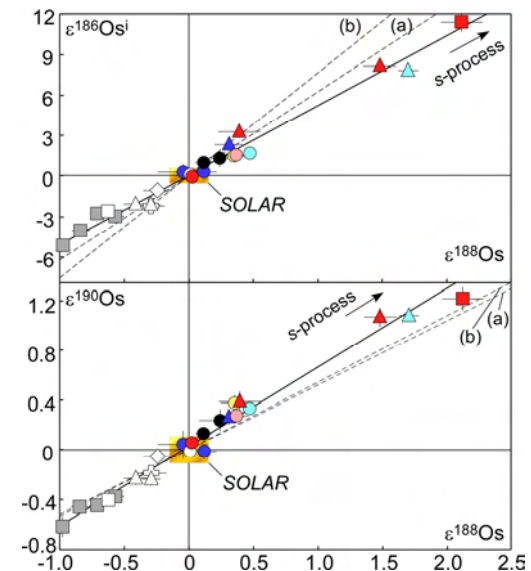

Fig. 2. $\varepsilon^{186}Os^i$-$\varepsilon^{188}Os$ and $\varepsilon^{190}Os$-^{188}Os plots for bulk carbonaceous chondrites and acid-resistant phases. Bold lines indicate the regressions for all chondrite samples. Dashed lines show mixing between the solar value and the s-process nucleosynthetic components calculated by the "stellar model" (a) and the "classical model" (b) of [12].