ORIGIN OF NUCLEOSYNTHETIC ISOTOPE HETEROGENEITY IN THE SOLAR NEBULA INFERRED FROM Mo AND W ISOTOPES IN ACID LEACHATES FROM MURCHISON. C. Burkhardt[1], T. Kleine[2], N. Dauphas[3] and R.Wieler[1]. [1]Institute of Geochemistry and Petrology, Clausiusstrasse 25, ETH Zürich NW D84, CH-8092 Zürich (burkhardt@erdw.ethz.ch). [2]Institut für Planetologie, Westfälische Wilhelms-Universität Münster, Wilhelm-Klemm Strasse 10, D-48149 Münster. [3]Origins Laboratory, Dept. of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, 5734 South Ellis Avenue, Chicago, IL 60637.

Introduction: Bulk chondrites and differentiated meteorites exhibit nucleosynthetic isotope anomalies at the bulk meteorite scale for a number of elements including Cr, Ti, Mo, Ru [e.g 1-3]. These anomalies are interpreted to indicate a large-scale heterogeneous distribution of isotopically diverse presolar material within the solar nebula. However, the origin of this heterogeneous distribution is poorly understood and existing models can be broadly subdivided into three groups. In the first group of models the heterogeneous distribution of presolar dust is a primordial feature of the solar nebula, either because it was inherited from the protosolar molecular cloud core [4] or because of a late injection of freshly synthesized matter into the disk [5]. In a second group of models physical sorting [6] or selective, thermal destruction [3] of presolar grains imparted a heterogeneous distribution of presolar dust in an initially well-mixed disk. Finally, a third model argues that at least for some elements a selective destruction of isotopically diverse carrier phases by parent body processes might have been important [7].

In spite of the isotopic heterogeneity documented for some elements, isotopic homogeneity exists for other elements, including Hf [8], Os [9], and, with the possible exception of IVB irons [10], also W [11]. Understanding why isotopic heterogeneities at the bulk meteorite scale exist for some elements but not for others is key for constraining the origin of the isotopic heterogeneity in the solar nebula.

To address this issue and to obtain new information regarding the presence of isotopically diverse presolar carriers of W, Mo and Os in the solar nebula we obtained the first W isotopic data for acid leachates of the Murchison carbonaceous chondrite. Mo isotopic data for the very same leachates were presented by us last year [12], and Os isotopic data for these leachates were reported in [13]. The new W isotope data in conjunction with the Mo and Os isotopic data are used to systematically explore the causes for the observed planetary-scale isotopic heterogeneity in some elements and their absence in others. Furthermore, the W isotopic data have important implications for the use of the short-lived ^{182}Hf-^{182}W system as a chronometer for early solar system processes.

Methods: The differential dissolution of Murchison was performed on ~16.5 g sample powder at the Univ. of Chicago using the following sequence [13]:

L1: 9 M HAc, 1 day, 20 °C;
L2: 4.7 M HNO_3, 5 days, 20 °C;
L3: 5.5 M HCl, 1 day, 75 °C;
L4: 13 M HF – 3 M HCl, 1 day, 75 °C;
L5: 13 M HF – 6 M HCl, 3 days, 150 °C;
L6: insoluble residue.

After drying down, aliquots of these samples were treated with aqua regia at ETH Zürich, dried and then redissolved in HCl. The insoluble residue (L6) was fused by a CO_2 laser under a reducing atmosphere and then digested in HNO_3-HF-$HClO_4$. For all six samples, Hf and W concentrations were determined by isotope dilution on small aliquots. Purification of W from the sample matrix followed our previously established techniques [14]. W isotopic compositions were determined using a Nu Plasma MC-ICP-MS at ETH Zürich and are reported in ε^iW values as the deviation from terrestrial W in parts per 10,000. Instrumental mass bias was corrected relative to ^{186}W/^{184}W=0.92767 using the exponential law. Isobaric interferences of Hf, Ta and Os on W isotopes were except for Hf and in some cases Os negligible. Due to the large Hf corrections and low ^{180}W concentrations, no meaningful data could be obtained for this isotope.

Results: The new W isotopic data for the different acid leachates (L1-L5) and the insoluble residue (L6) are shown in Figs. 1 and 2. All samples have W isotopic compositions different from the terrestrial value (Fig. 1), and the observed isotopic patterns are consistent with variable abundances of either s-process or r-process isotopes of W, as calculated using the s-process abundances of the stellar model [15] (Fig. 2).

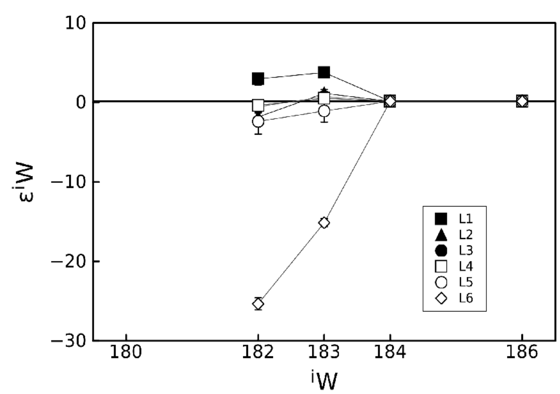

Fig. 1: W isotopic compositions of Murchison leachates (L1-L5) and an acid-resistant residue (L6) in an ^{i}W vs. ε^iW plot. Note that no data for ^{180}W are reported.

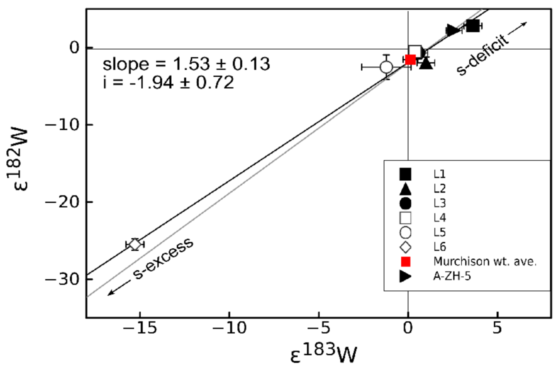

Fig. 2: Plot of $\varepsilon^{182}W$ vs. $\varepsilon^{183}W$. Calculated regression line of the data is similar to the s-process mixing line from the stellar model [15].

The weighted average of samples L1 to L6 yields ~120 ppb W, $\varepsilon^{182}W$ = -1.56 ±0.57, and $\varepsilon^{183}W$ = 0.12±0.40, consistent with previously reported data for bulk samples of Murchison [e.g., 11].

Discussion: *Implicatios for Hf-W chronometry.* Fig. 2 illustrates that there is good agreement between the linear regression of the W isotopic data and the predictions of the stellar model of s-process nucleosynthesis [15], indicating that the leachate data can be explained by the presence of at least two distinct carrier phases with variable W isotopic compositions (one enriched and one depleted in s-process W). The presence of $\varepsilon^{182}W$ variations in the leachates implies that not all ^{182}W variations in meteorites have chronological significance, such that the application of Hf-W chronometry requires monitoring the non-radiogenic W isotopes (e.g., $\varepsilon^{183}W$) to correct for potential nucleosynthetic anomalies. Such corrections are possible using the $\varepsilon^{182}W$-$\varepsilon^{183}W$ co-variation constrained by the stellar model and the leachate data (Fig. 2). It is noteworthy that the leachate data (and the stellar model [15]) predict larger nucleosynthetic ^{182}W anomalies for a given ^{183}W anomaly then the model used so far [10]. As a consequence, small $\varepsilon^{183}W$ anomalies in CAI [14], which were previously considered to have no significant effect on their $\varepsilon^{182}W$, now require a downward correction of the initial $\varepsilon^{182}W$ of CAI from ~ –3.3 $\varepsilon^{182}W$ [14] to ~ –3.5 $\varepsilon^{182}W$.

Origin of isotopic heterogeneity in the solar nebula. Fig. 3 shows that the W isotopic anomalies in the leachates are well correlated with those in Mo. This correlation also is in reasonable agreement with predictions of the stellar model of s-process nucleosynthesis. It is noteworthy that A-ZH-5, a fine-grained CAI with large nucleosynthetic Mo and W anomalies, also plots on the correlation line defined by the leachates.

The Mo and W leachate correlation can thus be used to calculate the expected nucleosynthetic W isotope anomaly for bulk meteorites, given their Mo isotope anomalies. Such a calculation reveals that resolvable nucleosynthetic W isotope anomalies should exist in bulk meteorites. This, however, is not observed (Fig. 3 inlet). Note that the small ^{183}W anomaly reported for IVB irons [10] is about a factor of two smaller than the anomaly that would be expected based on the Mo isotope composition of IVB iron meteorites. Thus, although the Mo and W isotopic anomalies are well correlated in acid leachates of primitive chondrites, they are decoupled at the bulk meteorite scale.

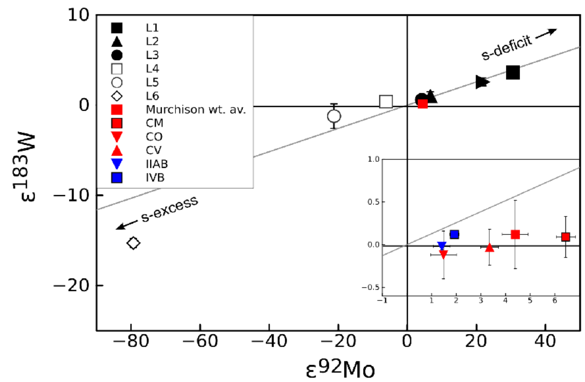

Figure 3: W and Mo isotopic anomalies in Murchison leachates correlate roughly as expected from nucleosynthetic theory [15], indicating that the anomalies are hosted in the same carrier(s). Bulk meteorite data, however, do not follow this correlation (small inlet).

This decoupling excludes that isotopic anomalies at the bulk meteorite scale (at least for Mo) are related to a primordial heterogeneous distribution of presolar dust within the solar nebula, but requires this isotopic heterogeneity to be generated by processes within an isotopically homogeneous nebula. As evident from strong depletions of Mo in CAI, Mo might be preferentially lost over W during evaporation under oxidizing conditions in the early solar nebula [16]. If such processes acted on presolar dust in the solar nebula, isotopically anomalous Mo from presolar components might have been preferentially lost with the vapor phase, leaving behind a residue having complementary Mo isotope anomalies but no (or smaller) W isotope anomalies. This model may also be capable of explaining the absence of Os isotope anomalies at the bulk meteorite scale, because under oxidizing conditions Os does not become volatile. Examining as to whether this new model is applicable to other elements will be an important task of future research.

References: [1] Burkhardt C. et al. (2011) *EPSL, 312,* 390-400. [2] Chen J. et al. (2010) *GCA, 74,* 3851-3862. [3] Trinquier et al. (2009) *Science, 324,* 374-376 [4] Clayton, D, (1982) *QJRAS, 23,* 174. [5] Bizzarro M. et al. (2007) *Science, 316,* 1178. [6] Dauphas N. et al. (2010) *ApJ, 720,* 1577. [7] Yokoyama et al. (2011) *EPSL, 305*, 115-123. [8] Sprung et al. (2010) *EPSL 295*, 1-11. [9] Yokoyama et al. (2010) *EPSL 291*, 48-59. [10] Qin et al. (2008) *APJ, 674*, 1234-1241. [11] Kleine et al. (2004) *GCA 68*, 2935-2946. [12] Burkhardt et al. (2011) *LPSC 42*, #2592. [13] Reisberg L. et al. (2009) *EPSL, 277*, 334-344. [14] Burkhardt et al. (2008) *GCA, 72*, 6177-6197. [15] Arlandini, C. et al. (1999) *ApJ, 525*, 886-900 [16] Fegley, B. and Palme, H. (1985) *EPSL 72*, 311-126.