$^{238}U/^{235}U$ VARIATIONS IN CAIs: IMPLICATIONS FOR PB-PB DATING. G.A. Brennecka[1], S. Weyer[2], M. Wadhwa[1], P.E. Janney[1], A.D. Anbar[1], [1]Arizona State University, Tempe, AZ, USA (*brennecka@asu.edu) [2]Institut fur Geowissenschaften, Universitaet Frankfurt, Germany

Introduction: It is generally believed that Calcium-Aluminum-rich inclusions (CAIs) were the first solids to condense from the cooling protoplanetary disk during the birth of our solar system [1], and the high-precision Pb-Pb dates obtained by various workers for CAIs have been considered the starting point of the Solar System for decades.

It has long been thought that $^{238}U/^{235}U$ is unfractionated in meteoritic material (=137.88 [2]), and this assumption is a cornerstone of the high precision geochronology that has defined the absolute ages of early Solar System materials. In this study, we present data obtained from CAIs of the Allende meteorite challenging the assumed uniformity of $^{238}U/^{235}U$ used in the Pb-Pb age equation for meteoritic material. Such data may require revision of the absolute age of the first solids in the Solar System.

Background: Recent measurements have shown $^{238}U/^{235}U$ fractionated by as much as ~1.3‰ on Earth [3], [4], [5]. This fractionation occurs mainly during low temperature redox transformations ($U^{IV} \leftrightarrow U^{VI}$) [5], [6]. The discovery of variable $^{238}U/^{235}U$ on Earth suggests $^{238}U/^{235}U$ might not be constant in other Solar System materials, and hence should be revisited using modern techniques and instrumentation.

Samples and Methods: Five CAIs were extracted from fragments of the Allende meteorite from the collection at the Center for Meteorite Studies at Arizona State University (ASU). The largest sample (CAI 165) was split after dissolution and measured in replicate. A section of bulk rock was also obtained for measurement. After the samples were crushed, rinsed and dissolved, they were prepared for measurement following the procedures outlined in Weyer et al. (2008).

Uranium isotope measurements were performed on a ThermoFinnigan Neptune MC-ICP-MS instrument at ASU, using a combination of amplifiers employing 10^{11} and 10^{12} Ohm resistors. A ^{236}U-^{233}U double spike was used to correct for instrumental mass bias during measurement. Standards were measured at the same approximate concentration as the samples, and sample errors (±2σ) are estimated from multiple runs of the standard during the same session as sample measurement.

Results: Results (Table 1; Figure 1) are reported relative to SRM-950a, a uranium standard used to define $^{238}U/^{235}U$ at 137.88 [4]. CAIs from Allende are variable with respect to the $^{238}U/^{235}U$ ratio. Two of the samples (including the CAI 165 duplicate) show a slight enrichment in ^{235}U with a $^{238}U/^{235}U$ of 137.839±0.012. One sample is unfractionated outside of analytical error. Samples 166 and 167 (the only Type A CAIs investigated) have not yet been measured for U isotopes due to their small size and extremely low U contents.

Sample	CAI Type	Weight (g)	Total ng U	$^{238}U/^{235}U$ (±0.012)
CAI 164	B	0.7048	~68	137.839
CAI 165	B	2.8383	~210	137.843
CAI 165a	B	Replicate	Replicate	137.835
CAI 166	A	0.1730	~3	n/a
CAI 167	A	0.3679	~9	n/a
CAI 168	B	1.3429	~78	137.885

Table 1 – Results from the CAIs of this study

Measurements of bulk Allende produce a $^{238}U/^{235}U$ of 137.818±0.012, well outside of analytical error from the "natural" ratio of 137.88. Results are complied in Figure 1.

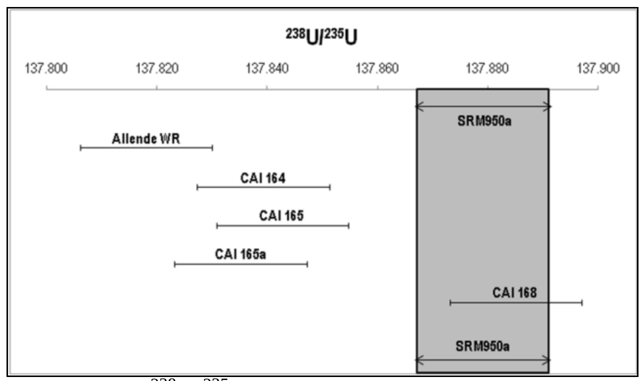

Figure 1 – $^{238}U/^{235}U$ ratios and errors from CAIs of this study. The shaded box represents the error calculated (±2σ) based on multiple measurements of the standard.

Discussion and Implications: The dependence of Pb-Pb ages on a precise knowledge of $^{238}U/^{235}U$ means that slight variations in $^{238}U/^{235}U$ require adjustment to the calculated ages of samples shown to deviate from the "natural" uranium isotope ratio of 137.88. Age adjustments for early Solar System materials depend on the magnitude and direction of the shift in $^{238}U/^{235}U$ relative to the canonical value, or 137.88 (Figure 2). The CAI measurements reported here translate to Pb-Pb age adjustment of approximately -0.4 Ma, making these samples slightly younger than previously thought.

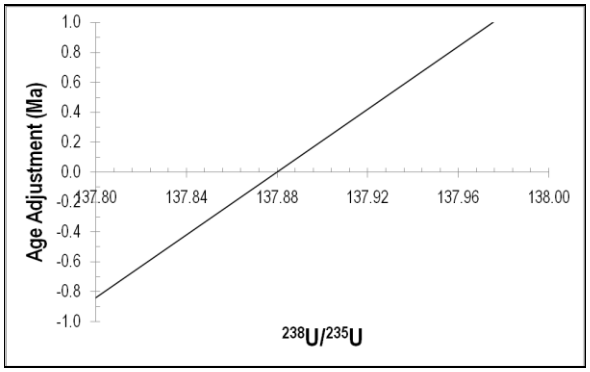

Figure 2 – Age adjustment based on different $^{238}U/^{235}U$ values

At this time, it is unclear if the variations of ^{235}U in these samples result from fractionation in a variable redox environment (as on Earth) or caused by inhomogeneous mixing and subsequent decay of "live" ^{247}Cm present during formation of the CAIs. It will be possible to differentiate between the mechanisms once more samples are studied to determine patterns and extent of variation in Solar System material.

While is beyond the scope of this study to definitively determine which possible mechanism caused the variation of ^{235}U in the CAIs of Allende, it is clear from these samples that $^{238}U/^{235}U$ can no longer be treated as a constant in Solar System material. The Pb-Pb dating technique is the only absolute dating technique able to resolve < 1 Ma differences in materials from the early Solar System. In order to produce robust Pb-Pb ages given the sensitivity of modern mass spectrometers, it is now essential to also measure the $^{238}U/^{235}U$ ratio in dated material.

References: [1] Grey et al. (1973) *Icarus*, *20*, 213-239. [2] Chen and Wasserburg (1980) *Geophysical Research Letters*, *7*, 275-278 [3] Stirling et al. (2007) *EPSL*, *264*, 208-225 [4] Weyer et al. (2008) *GCA*, *72*, 345-359 [5] Schauble (2006) *AGU Fall meeting abstract* [6] Brennecka et al. (2009) *In Prep*