

**BARIUM ISOTOPE ABUNDANCES IN METEORITES: IMPLICATIONS FOR EARLY SOLAR SYSTEM EVOLUTION** K.R. Bermingham<sup>1,2</sup>, K. Mezger<sup>1,3</sup>, E.E. Scherer<sup>1</sup>, R. Carlson<sup>4</sup>, M. Horan<sup>4</sup>, D. Upadhyay<sup>1,5</sup>, T. Magna<sup>1,6</sup>, A. Pack<sup>7</sup>. <sup>1</sup>Institut für Mineralogie, Westfälische Wilhelms-Universität, Corrensstraße 24, 48149 Münster, Germany; <sup>2</sup>Isotope Geochemistry Laboratory, Department of Geology, University of Maryland, College Park, MD-20740 USA (kberming@umd.edu); <sup>3</sup>Institut für Geologie, Universität Bern, Baltzerstrasse 1 + 3, 3012 Bern, Switzerland; <sup>4</sup>Department of Terrestrial Magnetism, Carnegie Institution of Washington, 5241 Broad Branch Road NW, Washington DC 20015 USA; <sup>5</sup>Indian Institute of Technology Kharagpur, 721302, Kharagpur, India; <sup>6</sup>Czech Geological Survey, Klárov 3, 11821 Prague 1, Czech Republic; <sup>7</sup>Geowissenschaftliches Zentrum, Georg-August-Universität, Goldschmidtstraße 1, 37077 Göttingen, Germany.

**Introduction:** The origin and degree of isotopic heterogeneity in the early Solar System are major unknowns in planetary science. Various nucleosynthetic processes contributed to the abundances of the elements and their isotopes in the Solar System. How these nucleosynthetic materials were mixed together to form the Solar nebula is currently unclear. Some elements display anomalies at the “whole rock” meteorite scale in one or more isotopes (<sup>46,50</sup>Ti [1,2], <sup>54</sup>Cr [3,4], <sup>62</sup>Ni [5,6], <sup>92,94,95,97,100</sup>Mo [7], <sup>100</sup>Ru [8], <sup>135,137</sup>Ba [9-12], <sup>142</sup>Nd, <sup>144</sup>Sm [11,12], <sup>180,184</sup>W [13,14,15]), whereas others have no detectable isotope anomalies (Os [16], Te [17]). Here, the distribution of Ba isotopes in whole rock meteorites (eucrites, diogenites, ordinary chondrites, Martian meteorites, enstatite- and carbonaceous chondrites) was determined to constrain the levels of Ba isotope heterogeneity on the regional scale in the Solar System.

**Methods:** Traditionally, whole rock isotopic analyses of low metamorphic grade meteorites have been difficult because of the presence of acid-insoluble components with extreme isotopic compositions (e.g., presolar SiC [12]). A melting method using aerodynamic levitation combined with CO<sub>2</sub>-laser heating was used in our study to oxidize the SiC [18]. All low metamorphic grade meteorites were melted by heating ~10 mg of the powdered sample for 5 - 10 s, multiple times. The resulting melt beads were placed in an ultrasonic bath for 10 minutes to remove adhering material. The beads were then digested using a concentrated HF-HNO<sub>3</sub> (3:1) mixture, achieving complete sample dissolution. Any samples containing white precipitates, presumably fluorides, were dried down repeatedly with HClO<sub>4</sub> until the sample formed a clear solution when dissolved in 6 M HCl.

A chemical separation method derived from previous studies was used to separate and isolate Ba, REE, and the major elements [11,12,19]. Isotope measurements were performed on the Thermo-Finnigan Triton thermal ionization mass spectrometer (TIMS) at the Zentrallabor für Geochronologie, Universität Münster, Germany. All data presented here have been corrected for mass fractionation using the exponential law with the reference ratio <sup>134/138</sup>Ba and are reported with 2σ<sub>m</sub> uncertainties. The data are

presented using the μ notation (deviations in parts per 10<sup>6</sup> from the terrestrial standard).

**Results:** Barium isotope ratios for all analyzed meteorite samples fall within the external reproducibility of the terrestrial Ba standard solution (Figure 1). The absence of Ba isotope deviations from the terrestrial composition implies that Ba was well mixed on a regional scale in the Solar nebula. The Ba isotope compositions of equilibrated meteorites (e.g., eucrites and ordinary chondrites of high metamorphic grade) agree with literature data on the same kinds of meteorites as well as the terrestrial values. However, the Ba isotope compositions of the primitive, thermally unequilibrated samples, which also fall within terrestrial isotope compositions, do not agree with published Ba isotope data from the same type of meteorites (e.g., CM2 and EH4 [9,11,12]). A possible reason for this discrepancy may be incomplete digestions in the previous studies. Several carbonaceous chondrites are currently being processed for Ba isotope composition to further investigate the suggested terrestrial Ba isotope composition of thermally unequilibrated meteorites.

**Discussion:** The present results show that Ba isotopes, and thus their carrier phases, were homogeneously distributed in the Solar System (i.e., within the analytical uncertainties of ~0.2 ε units for r- and s-process isotopes and ~2 ε units for p-process isotopes) throughout the feeding zones for all meteorite types analyzed.

Barium and other elements that were found to have homogeneously distributed isotope compositions (Te and Os) are in contrast to isotopically anomalous elements (Ti, Cr, Ni, Mo, Ru, Nd, Sm, and W) from thermally equilibrated and unequilibrated meteorites. There appears to be a correlation between the number of nucleosynthetic source(s) of an element and the distribution of its isotopes in the Solar System. Anomalies seem to be essentially undetectable if an element is dominated by one nucleosynthetic pathway. However, if an element comprises isotopes that have roughly equal proportions of s- and r-process nuclides, then small deviations in isotope abundances at the ε-level on the bulk sample scale may be detected. These anomalies stem from slight variations in the distribution of carrier mineral grains that have isotope

anomalies on the ‰-level [20]. These relationships, in conjunction with incomplete homogenization of the solar nebula with respect to carrier grain distribution, can explain the coexistence of isotopically heterogeneous and homogeneous elements in the early Solar System.

As analytical precision continues to improve, isotope anomalies in what are currently classified as “homogeneously” distributed isotopes will probably be detected as suggested by higher precision data for Ba [11,12]. Because the vast majority of Ba (~80% [21]), is derived from a single nucleosynthetic pathway, the s-process, it may be difficult to resolve nucleosynthetic anomalies in Ba. If nuclear anomalies are present, however, they would be expected in  $^{135}\text{Ba}$  and  $^{137}\text{Ba}$ , because both have a substantial r-process component, and  $^{130}\text{Ba}$  and  $^{132}\text{Ba}$  (p-process).

**Conclusions:** The small (‰-level) to absent isotope variations on the bulk meteorite scale implies that the solar nebula was chemically and physically fairly well mixed. Regarding chronometry, for those parent and reference isotope pairs that are single-process-dominated, such that all materials have essentially the same initial parent-reference isotope ratio, a major assumption of short-lived chronometry holds. Chronometers that comprise parent and reference isotopes produced by a balance of different processes (s, p, ± r), resulting in epsilon-level isotope ratio anomalies, should nevertheless be robust because percent-scale variations in heterogeneity are required before the age errors become analytically significant.

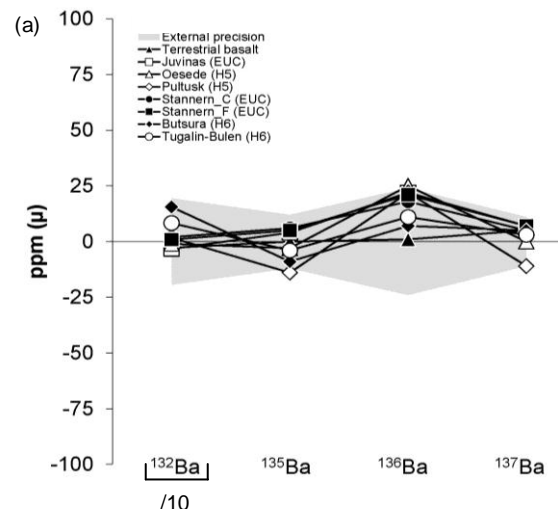
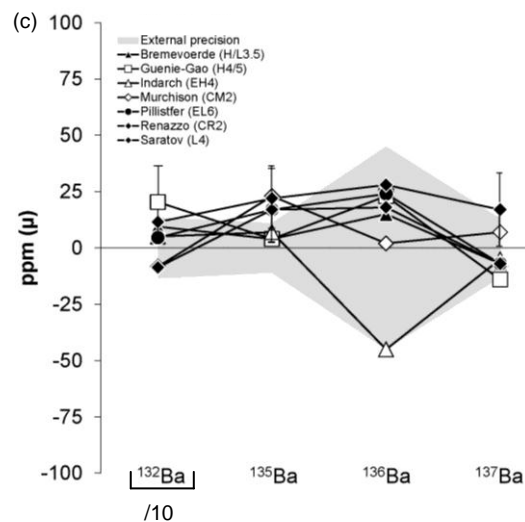
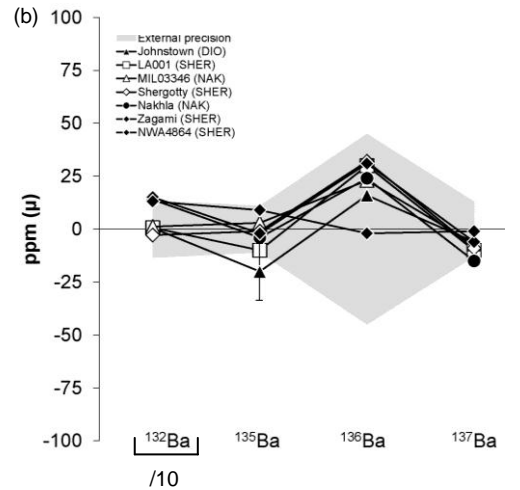


Figure 1.

The average Ba isotope ratios (in ppm) of individual filament measurements of (a) thermally equilibrated meteorites; (b) Martian meteorites; and (c) thermally unequilibrated meteorites. Both the values and uncertainties of  $^{132}\text{Ba}$  have been divided by 10 to plot on this scale. The reported external precision varies between (a) and (b,c) because the standards used to calculate these errors were analyzed during different measuring campaigns.



## References:

- [1] Leya et al., 2008. *Earth Planet. Sci. Lett.* 266, 233-244.
- [2] Trinquier et al., 2009. *Science* 324, 374-376.
- [3] Qin et al., 2010. *Geochim. Cosmochim. Acta* 74, 1122-1145.
- [4] Yamakawa et al., 2010. *Ap. J.* 720, 150-154.
- [5] Regelous et al., 2008. *Earth Planet. Sci. Lett.* 272, 330-338.
- [6] Quitté et al., 2010. *Ap. J.* 720, 1215-1224.
- [7] Burkhardt et al., 2011. *Earth Planet. Sci. Lett.* 312, 390-400.
- [8] Chen et al., 2010. *Geochim. Cosmochim. Acta* 74, 3851-3862.
- [9] Hidaka et al., 2003. *Earth Planet. Sci. Lett.* 214, 455-466.
- [10] Ranen and Jacobsen, 2006. *Science* 314, 809-812.
- [11] Andreasen and Sharma, 2007. *Ap. J.* 665, 874-883.
- [12] Carlson et al., 2007. *Science* 316, 1175-1178.
- [13] Qin et al., 2008. *Ap. J.* 674, 1234-1241.
- [14] Irisawa et al., 2009. *Geochem. J.* 43, 395-402.
- [15] Schulz and Münker, 2011. *Min. Mag.* 75, 1827.
- [16] Walker, 2012. *Earth Planet. Sci. Lett.* 351-352, 36-44.
- [17] Fehr et al., 2009. *Meteoritics Planet. Sci.* 44, 971-984.
- [18] Pack et al., 2010. *Geochem. Trans.* 11, 4-16.
- [19] Eugster et al., 1969. *J. Geophys. Res.* 74, 3897-3908.
- [20] Marhas et al., 2007. *Meteoritics Planet. Sci.* 42, 1077-1101.
- [21] Arlandini et al., 1999. *Ap. J.* 525, 886-900.