Ba, Nd AND Sm ISOTOPE ANOMALIES IN MURCHISON LEACHES: DISTINCT CARRIERS OF S- AND R-PROCESS NUCLEOSYNTHETIC COMPONENTS  R.W. Carlson, L. Qin, and C. M. O’D. Alexander, Department of Terrestrial Magnetism, Carnegie Institution of Washington, 5241 Broad Branch Road, NW, Washington, DC 20015, USA (E-mail: rcarlson@ciw.edu).

Introduction: Recent high-precision analyses have revealed isotope anomalies in Ba, Nd and Sm at the whole rock scale in carbonaceous chondrites [1-4]. The Ba anomalies are readily interpreted as reflecting an excess of r-process over s-process produced isotopes in the whole rocks [1,3]. Sm contains resolved p-process deficiencies [2], but neither Sm nor Nd show clearly resolved s-r-process anomalies [1,2]. Hidaka et al. [5] showed that the anomalies in Ba could be enhanced by factors of as much as 4 over the whole rock data by mild leaching. We report here the results of similar leaching experiments of Murchison. Comparison of the magnitude and cause of isotopic variation in Ba, Nd and Sm can provide information on: 1) the relative abundances of the isotopes of these elements that can be compared with nucleosynthetic models for their genesis, 2) the nature of the carriers of the isotope anomalies and whether they are the same for Ba and the REE, and 3) the consequences of nucleosynthetically-induced isotope variations for the 140Ba-142Nd radiometric system.

Methods: Five grams of Murchison powder were sequentially leached in: 1) 8.5N acetic acid at room temperature for 24 hours, 2) 6N HNO₃ at room temperature for 5 days, and 3) 6N HCl at 75°C for 24 hours. Half of the residue remaining after these leaching steps was then dissolved completely in a mixture of concentrated HNO₃ and HF in a Parr bomb held at 170°C for a week. The leaches and dissolved residue were processed through a series of ion exchange columns [1] that resulted in the isolation of clean Ba, Nd and Sm splits. The isotopic composition of these elements was then determined on the DTM Triton thermal ionization mass spectrometer using procedures similar to those described previously [1].

Results and Discussion: The data for Ba in the leaches reproduces the results of Hidaka et al. [5] quite well. Most leaches show excesses in the r-process isotopes 135Ba and 137Ba (Fig. 1). Our data include the p-process isotopes 130Ba and 132Ba that are elevated in the leaches by 2 to 4 parts in 10,000 in comparison to the terrestrial Ba standard. The residue data show the same complimentary deficiencies in 135Ba and 137Ba found by [5], but the anomalies found here are nearly 50 times larger and include large deficits in 130Ba and 132Ba. The anomalies in the residue are well modeled as a 1.12% excess in pure s-process Ba of the isotopic composition given by [6]. The largest of the leach anomalies are reasonably well fit, with some mismatch at 137Ba and 138Ba, by assuming a 0.029% deficiency in the s-process component. The results provide no clear evidence for variable p-process contributions as the anomalies found in 138Ba and 132Ba can be matched simply by varying the ratio of r- and s-process Ba, at least within the resolution of the models and data.

![Figure 1: Ba isotope data normalized to the 2 s-only isotopes 134Ba/136Ba. The data are shown as parts in 10,000 (ε) deviation of each ratio compared to the terrestrial standard. Dotted lines show the patterns created by adding or subtracting pure s-process Ba [6] to the terrestrial standard until the patterns match the magnitude of the 135Ba anomaly.](image)
that s-process enriched pre-solar SiC may contribute substantially to the residue Ba, but not the REE, budget. The Sm anomalies in leach 1 require a slightly smaller s-process deficit (0.015%) than the best fit for Ba when forced to fit the data for $^{144}\text{Sm}$, but the s-deficit patterns overpredict the size of the anomalies in $^{152}\text{Sm}$ and $^{154}\text{Sm}$. Hoppe and Ott [6] do not report data for $^{154}\text{Sm}$, so the calculations shown here assume no s-process production of $^{154}\text{Sm}$. For the residue, the predicted anomalies in $^{152}\text{Sm}$ and $^{154}\text{Sm}$ are smaller than observed. The $^{149}\text{Sm}/^{154}\text{Sm}$ ratio reported for Murchison whole rock by [2] is only 0.18$\varepsilon$ lower than the terrestrial standard. Our data are consistent with this observation and indicate that thermal neutron capture effects, which can modify the abundance of $^{149}\text{Sm}$ and $^{150}\text{Sm}$, are minimal in Murchison. Murchison whole rock shows a deficit in $^{144}\text{Sm}$ of 0.96$\varepsilon$. If this deficit is due to an s-excess, $^{149}\text{Sm}/^{154}\text{Sm}$ should be high by 0.48$\varepsilon$. This excess is not observed [2], which supports the interpretation that the low $^{144}\text{Sm}$ is a p-process deficit [2]. The p-process anomaly, however, is lost in the magnitude and uncertainty of the s-, r- contribution to $^{146}\text{Sm}/^{148}\text{Sm}$ in the data reported here.

Nd shows similar magnitude and sign anomalies as Sm in both leaches and residue (Fig. 3). An s-excess of 0.098% matches the pattern and magnitude of the anomalies seen in the residue while a 0.05% s-deficiency is needed to match the largest leach anomalies. Measured anomalies in $^{143}\text{Nd}$ and $^{144}\text{Nd}$ are correlated (Fig. 4), suggestive of a contribution from Sm decay. The slope of the correlation, however, corresponds to a $^{146}\text{Sm}/^{144}\text{Sm}$ ratio equal to about 10 times the initial solar system value of 0.008. Correcting for the unequal s-, r-process distribution moves the data closer to a slope consistent with the initial $^{146}\text{Sm}/^{144}\text{Sm}$ of the solar system. To explain the circa 0.2$\varepsilon$ difference in $^{144}\text{Nd}/^{144}\text{Nd}$ between ordinary chondrites, eucrites and modern terrestrial rocks by an s-process excess in the Earth requires only a 0.004% excess in s-process Nd. This magnitude of s-, r-process variation would leave anomalies of 0.009:0.23:0.27$\varepsilon$ in $^{145}\text{Nd}$,$^{145}\text{Nd}$,$^{150}\text{Nd}$, respectively. Anomalies of this magnitude approach measurement precision with current techniques. The whole rock chondrite data reported by [1] shows an average $\varepsilon^{150}\text{Nd}$ of 0.03 ± 0.24. No resolvable correlation is observed between $^{142}\text{Nd}$,$^{144}\text{Nd}$ and $^{150}\text{Nd}/^{144}\text{Nd}$ in whole rock chondrites leaving $^{146}\text{Sm}$ decay and a non-chondritic Sm/Nd ratio as the likely explanation for the elevated $^{142}\text{Nd}$,$^{144}\text{Nd}$ in modern terrestrial Nd.