Decoupling of Barium and Lanthanide r-process nuclide sources: constraints on the origin of terrestrial ^{142}Nd anomalies. Rasmus Andreasen and Mukul. Sharma, Radiogenic Isotope Geochemistry Laboratory, Department of Earth Sciences, Dartmouth College, 6105 Sherman Fairchild Hall, Hanover NH 03755, USA (e-mail: rasmus.andreasen@dartmouth.edu, mukul.sharma@dartmouth.edu)

Introduction: Recent high-precision studies of small but resolvable variations in the isotopic composition of Sm, Nd [1,2,3] and Ba [4] in bulk chondritic meteorites have yielded conflicting results regarding the evolution of early Earth. The key issue is whether the observed variations in ^{142}Nd between the terrestrial samples and other planetary bodies are due to different contributions of nucleosynthetic (p-, s-, and r-process) components or they are caused by differences in Sm/Nd ratios generated through planetary differentiation while short-lived ^{146}Sm (^{146}Sm→^{142}Nd; $t_{½}$ = 103 Ma) was alive. That the terrestrial upper mantle displays a ^{142}Nd anomaly of around +18±8 μ (=1 part in 10^6) with respect to chondrites and basaltic eucrites has been taken as evidence for global differentiation within 30 Ma after the formation of the Earth [1]. This inference is supported by the finding that the bulk moon Nd isotopic composition is chondritic [5]. However, as ^{142}Nd is an s-process nuclide with a small p-process contribution and the rest of the Nd isotopes have variable r-process contributions, the possibility remains that the chondrites and the eucrite parent bodies have an r-process excess (or p- or s-process deficit) relative to Earth. This question was investigated by [2] who discovered that carbonaceous chondrites have a deficit of about 100 μ in the p-process ^{144}Sm (and ^{146}Sm), which is consistent with and likely the cause of the bimodality in the ^{142}Nd values for carbonaceous versus ordinary chondrites (Fig. 1a). The ^{148}Sm/^{154}Sm and ^{145}Nd/^{144}Nd ratios in ordinary and carbonaceous chondrites, a eucrite and Earth indicate a Solar Nebula with a rather uniform r/s ratio. This strongly supports early differentiation as the cause for the elevated ^{142}Nd values seen in terrestrial samples relative to other planetary bodies. A different approach to the same question was taken by [4] who found anomalies in ^{137}Ba and ^{138}Ba in both carbonaceous and ordinary chondrites and attributed them to excesses in r-process or neutron burst material in the meteorites relative to Earth. Such an excess would produce a negative ^{142}Nd anomaly in the chondrites and thus the Ba isotope data do not require early terrestrial silicate differentiation. Here high-precision Ba and Sr isotopic compositions for the same sample aliquots (Allende CV3, Murchison CM2, Saint Severin LL6, and Juvinas) that were used for Sm and Nd isotope analyses are presented.

Methods: About 500 ng Sr or Ba were loaded on a Re side filament in 1 μl of 2.5 M HCl and 1 μl of 0.2 M H_3PO_4. Sample measurements were interleaved with standards runs. Sr isotopic compositions were measured in static mode at ^{88}Sr = 1×10^{-10} A. For Barium all seven isotopes were measured in static mode. For the Ba analyses a ^{138}Ba intensity of ~2×10^{-10} A was achieved for 4 hours or more. Isobaric interferences of La and Ce were < 0.1 ppm and negligible.

Results: If the p-process yields follow a scaling law [6], we expect ~100 ppm deficits in the p-nuclides ^{84}Sr, ^{130}Ba, and ^{132}Ba in carbonaceous chondrites. There are, however, no anomalies in ^{84}Sr in any of the analyzed meteorites, at the level of external reproducibility (±34 ppm, 2σ). In order to assess potential p-process deficits in ^{130}Ba and ^{132}Ba in carbonaceous chondrites and to substantiate the excesses in ^{137}Ba and ^{138}Ba in carbonaceous and ordinary chondrites as observed by [4], data were corrected for mass fractionation using ^{134}Ba/^{136}Ba, both s- only nuclides. The external reproducibilities of the ^{130}Ba/^{136}Ba and ^{132}Ba/^{136}Ba ratios are 55 ppm and 77 ppm (2σ), respectively. They are not sufficient to resolve the ~100 ppm negative anomalies in the p-isotopes expected for the carbonaceous chondrites. However, both Murchison and Allende show well resolved and identical anomalies of ~+39 μ and ~+22 μ for ^{135}Ba and ^{137}Ba, respectively (Fig. 1b). Significantly, the ^{138}Ba/^{136}Ba ratio of the carbonaceous chondrites cannot be resolved from the terrestrial mantle. These results are remarkable in that anomalies of the same magnitude (+39±7 μ in ^{135}Ba and +23±8 μ in ^{137}Ba) have been reported for Orgueil and Allende [7]. The Ba isotopic compositions of Saint Severin and Juvinas are indistinguishable from that of the terrestrial mantle. However, there is a clear discrepancy between these results and the ^{137}Ba and ^{138}Ba anomalies reported for Allende and Murchison by [4].

Discussion: The observed Ba anomalies in Allende and Murchison are consistent with an r- excess as calculated by [8]. In comparison to the carbonaceous chondrites, Saint Severin was subjected to early high metamorphism and Juvinas was produced from the melting and differentiation of an asteroid. These observations suggest that the Ba anomalies in carbonaceous chondrites could be an artifact of incomplete dissolution of acid resistant SiC grains from AGB stars that carry s-process Ba nuclides [9] and are destroyed during high degree of metamorphism or melting [10]. However, Allende and Murchison have vastly different SiC abundances suggesting that incomplete dissolution of presolar grains should result in essentially no Ba anomaly in Allende and pronounced anomalies in Murchison. Furthermore, high-precision Nd and Sm isotopic data from the same sample aliquots [2] do not show s- deficits, to be expected if the Ba isotopic patterns were produced from the incomplete dissolution of SiC grains. If an r- excess similar to that in Ba is present in Sr for the carbonaceous chondrites, the ^{84}Sr/^{86}Sr ratio would increase by ~12 μ, this effect is not large enough to mask a p-process deficit in ^{84}Sr of the magnitude of that seen for Sm and Nd. The absence of measurable deficits in ^{84}Sr for the carbonaceous chondrites is not due to a combination of p-deficit and r- excess.

Thus those isotopic ratios of Sr, Ba, Sm and Nd that are sensitive to variations in r/s ratios are quite uniform for ordinary chondrites, eucrite parent body, and Earth suggesting that s- and r-process nuclides were, on average, rather homogeneously distributed in the inner Solar Nebula (between 1 and 2.4 AU). This conclusion is consistent with an inferred uniform distribution of s- and r- process Mo [11], Ru [12], and Zr [13] isotopes in the inner solar nebula, and with recent simulations of terrestrial planet formation that suggest extensive radial mixing of the material between 1 and 2.5 AU (e.g., [14]). In contrast, both Allende and Murchison display deficits in p-process Sm and Nd isotopes [2] and excesses in r-process ^{135}Ba and ^{137}Ba. As the carbonaceous chondrites likely formed in the outer asteroidal belt (>2.7 AU) the p-, s, and r- nuclei were heterogeneously distributed in the Solar Nebula, in the regions >2.7 AU.

The r-process decoupling seen for Ba and REE in carbonaceous chondrites requires that either the r-sources for Ba and REE are different, or that the carrier phases of these elements have been differentially incorporated. The latter scenario is less likely as both Ba and REE should be associated with silicate phases. That there are distinctive r-sources with distinctions in the different source contributions below and above atomic mass ~140 is well established (e.g., [15,16]). The data presented here suggest a decoupling of r-process Ba from Nd and Sm. If so, the two types of r-sources have to be split at atomic number $Z \leq 56$ and $Z > 56$, in line with the assessment of [17].

The early terrestrial evolution model proposed in [1] is based on the small shifts in isotopic ratios due to radioactive decay and is strongly dependent on the initial Nd isotopic composition and a "bulk solar" Sm/Nd ratio of the Earth. In this study, we have shown that there are small but distinct differences in the isotopic composition of carbonaceous chondrite parent body on one hand and eucrite parent body, LL-chondrite parent body, and Earth on the other. That the latter display identical Sr, Ba, and Sm isotopic compositions are evidence that their initial Nd isotopic states were possibly identical (see also [2]). This still leaves the issue of whether different planetary bodies accreted with the same average bulk solar Sm/Nd ratio and remains to be fully resolved.

References: [1] Boyet M. & Carlson R. W. (2005) *Science, 309,* 576–581. [2] Andreasen R. & Sharma M. (2006) *Science, 314,* 806-809. [3] Boyet M. & Carlson R. W. (2006) *EPSL, 250,* 254–268. [4] Ranen M.C. & Jacobsen S. B. (2006) *Science, 314,* 809–812. [5] Rankenburg K. et al. (2006) *Science, 312,* 1369-1372. [6] Hayakawa T. et al. (2006) *Nucl.Phys.A, 758,* 525-528. [7] Harper C. L. et al. (1992) *Meteoritics, 27,* 230-231. [8] Arlandini C. et al. (1999) *AstrophysJ, 525,* 886-990. [9] Ott U. & Begemann F. (1990) *AstrophysJ, 353,* L57-L60. [10] Huss G. R. (1990) *Nature 347,* 159-162. [11] Becker H. & Walker R. J. (2003) *Nature 425*, 152-155. [12] Becker H. & Walker R. J. (2003) *ChemGeo 196*, 43-56. [13] Schönbachler M. et al. *EPSL, 216,* 467–481. [14] O'Brien et al. (2006) *Icarus, 184,* 39-58. [15] Sneden C. et al. (1996) *AstrophysJ, 467,* 819-840. [16] Wasserburg G. J. et al. (1996) *AstrophysJ, 466,* L109-L113. [17] Otsuki K. et al. (2003) *NewAstronomy, 8*, 767-776.

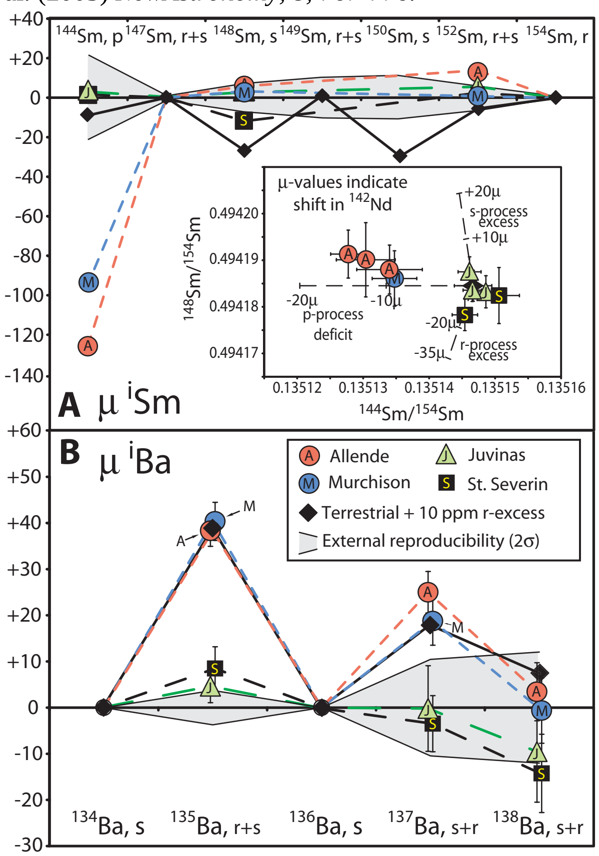

Figure 1.