

EXCESS 135 -BARIUM FROM LIVE 135 -CESIUM IN ORGUEIL CHEMICAL SEPARATES. R. H. Nichols, Jr., J. C. Brannon, and F. A. Podosek, Department of Earth and Planetary Sciences, Washington University, St. Louis, MO, 63130 (rhn@levee.wustl.edu).

Background: The first inference for the presence of live ^{135}Cs ($\tau_{1/2} = 2.3$ Ma) in the early solar system was based on a -1.8 ± 0.7 ϵ depletion (monoisotopic) in the $^{135}\text{Ba}/^{138}\text{Ba}$ ratio in the Allende FUN inclusion C1 [1], suggesting an initial $(^{135}\text{Cs}/^{133}\text{Cs})_i = (1.4 \pm 0.6) \times 10^{-4}$ for the non-FUN solar system composition (re-computed herein using recent solar system abundances [2]). From 1978 to 2000 subsequent analyses of meteoritic Ba exhibited excesses at ^{135}Ba , however, these were accompanied by correlated ^{137}Ba excesses, indicating an r-process/s-process enrichment in the analyzed samples (e.g. SiC-depleted) rather than evidence for the decay of live ^{135}Cs in the solar system [cf. 3]. The first direct evidence for the presence of live ^{135}Cs (i.e. excess ^{135}Ba correlated with Cs/Ba elemental ratios) was reported for acetic acid and HCl leachates of the Beardsley (H5) chondrite: $(^{135}\text{Cs}/^{133}\text{Cs})_i = (1.1 \pm 0.1) \times 10^{-4}$ [4]. Subsequent analyses of six Allende CAIs [5] suggest an initial $(^{135}\text{Cs}/^{133}\text{Cs})_i = (4.82 \pm 0.79) \times 10^{-4}$, significantly higher than [1, 4]. These measurements imply that aqueous alteration ceased on Beardsley (H5) and Zag (H3-6) 11.9 and 17.6 Ma, respectively, after CAI formation [5].

This Work: We have measured the Ba isotopic compositions in eleven sequential step-wise dissolution fractions from Orgueil (in particular the whole rock sample designated O-I [cf. 6,7,8]). These samples (as well as additional Orgueil, Murchison, Allende and Semarkona samples) have previously been analyzed for Cr [6, 7], K [8], and Ca, Fe, Zn, Rb and Sr [8], and have exhibited prominent isotopic excesses at ^{54}Cr [6,7] and ^{40}K [8], and minor excesses at ^{66}Zn [7]. A motivation for this work is to elucidate the origin and carriers of these anomalies, as well as identify other isotopic effects, in this case those which may be associated with Ba.

Results: The samples were analyzed by thermal ionization mass spectrometry (TIMS). Sample and standard (1 μg reagent) Ba in 1 N HCl were loaded on degassed and oxidized Ta filaments with phosphoric acid. Data were acquired on the Daly detector at beam intensities of 400 and 750 kcps ^{138}Ba (N.B. not Faraday). A single analysis is comprised of 2500 one second integrations per isotope (cycling through peaks at five 1 sec integrations), in addition to monitoring the baseline and masses 128, 139 and 140 for potential Te, La and Ce interferences (which were small or negligible relative to the errors dominated by Poisson statistics).

All data are referenced to ^{138}Ba and were initially corrected for instrumental discrimination based on $(^{135}\text{Ba}/^{138}\text{Ba}) = 0.09140$ [1], following standard TIMS procedure. In this normalization scheme, however, all ratios (except for the normalization ratio) for several samples (O-I-2,3,4,5) were depleted relative to standard composition. This situation is analogous to, but in the opposite direction of, the $^{135}\text{Ba}/^{138}\text{Ba}$ normalization scheme used by [1] for the Allende CAI C1. In the absence of any known process that could account for depletions in all isotopes except for ^{135}Ba , we re-normalize the data to ^{136}Ba (rather than ^{134}Ba , due to the higher analytic precision at ^{136}Ba ; similar results are obtained by $^{134,137}\text{Ba}$ normalizations). Under the $^{136}\text{Ba}/^{138}\text{Ba}$ normalization all ratios for these samples are isotopically normal (within 2σ error) except for the $(^{135}\text{Ba}/^{138}\text{Ba})$ ratios (Figure 1), which peak at $+7.6 \pm 3.8$ ϵ -units (2σ error) for sample O-I-3 (4 N HNO_3 dissolution at 25 $^\circ\text{C}$ for 5 days[5]).

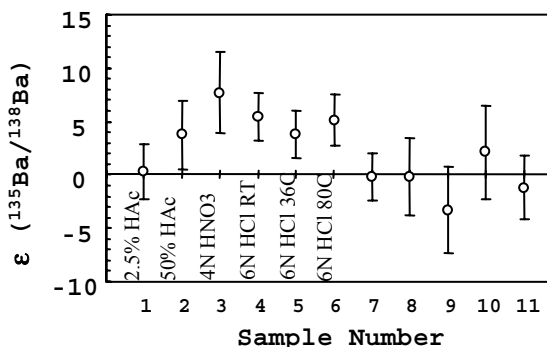


Figure 1. $(^{135}\text{Ba}/^{138}\text{Ba})$ deviations from normal in ϵ -units (2σ errors) for O-I stepwise dissolution fractions [6]. Data have been re-normalized to ^{136}Ba . All other isotopes are isotopically normal within error.

Discussion: The ^{135}Ba enrichments occur in the acetic acid, nitric acid and early HCl fractions, as may be expected from [4]. Previous measurements of Ba in bulk Orgueil [cf. 3] indicate an excess of 24 ± 8 ppm at $(^{135}\text{Ba}/^{136}\text{Ba})$ correlated with an excess of 16 ± 8 ppm at $(^{137}\text{Ba}/^{136}\text{Ba})$. No correlated excesses at ^{137}Ba are observed in the present dissolution fractions, nor would any be detected at the ppm level via ion counting. The mean ^{135}Ba excess in all eleven fractions (weighted by estimated Ba concentrations in each fraction; see below) is normal within 3σ , yet still consistent with the $^{135,137}\text{Ba}$ ppm-level excesses [cf. 3]. No discernable correlation exists between these enrich-

ments and any other isotopic anomalies previously measured in these dissolution fractions [5-7], nor would any be expected in most nucleosynthetic scenarios. While these excesses are most likely due to the decay of ^{135}Cs (invoking Occam), the appropriate question is when did the decay occur. Although representative of solar system composition [1], Orgueil is known to harbor a relatively high concentration of pristine presolar material (cf. [9]), much of which is probably yet undetected. Further, these specific Orgueil dissolution fractions also exhibit isotopic anomalies in other elements [6-8]. As such, these effects may be relict and carried by presolar material. To establish that the decay occurred in the early solar system we can, however, show a reasonable correlation between the bulk of the parent element Cs in Orgueil and daughter isotope ^{135}Ba .

The elemental abundances of Ba and Cs in the O-I fractions are unknown at the present time. We may, however, use the estimated elemental abundances of K or Rb [7,8] as a proxies for the abundance of Cs, and the known Mg, Ca or Sr abundances [6] as proxies for the abundance of Ba. A “traditional” 3-isotope isochron plot based on the proxies [K] and [Ca] is shown in Fig. 2. (The results are not significantly affected if other elements are chosen as proxies).

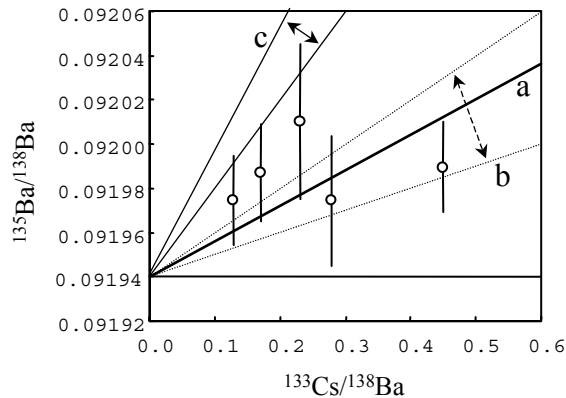


Figure 2. Cs-Ba isochron plot for samples O-I-2 through O-I-6. Abscissa values are computed using [K] and [Ca] abundances [6,7] as proxies for [Cs] and [Ba] abundances, respectively. Line “a” corresponds to the best fit through the data, $(^{135}\text{Cs}/^{133}\text{Cs})_I = 1.6 \times 10^{-4}$. The dashed range “b” corresponds to $(^{135}\text{Cs}/^{133}\text{Cs})_I = (1.4 \pm 0.6) \times 10^{-4}$, inferred by [1]. The range “c” corresponds to $(^{135}\text{Cs}/^{133}\text{Cs})_I = (4.8 \pm 0.8) \times 10^{-4}$, measured by [5].

The estimated Cs/Ba ratios are slightly higher than the chondritic (0.08 [2]), yet are much lower than the Cs/Ba ratios (~ 3 and 20) in the dissolution fractions measured by [4]. Although there is a large scatter in the data and these samples do not necessarily represent a single mineral phase, the data are consistent with previously determined initial $(^{135}\text{Cs}/^{133}\text{Cs})_I$ on the order of 10^{-4} . The inferred time difference between CAI formation (“c”) and Cs-Ba closure (perhaps cessation of aqueous alteration on the Orgueil parent body) in these carriers in Orgueil (“a”) is 3.6 Ma, consistent with the later compaction age of Orgueil, ~ 15 Ma [10].

References: [1] McCulloch M. T. and Wasserburg G. J. (1978) *Ap. J.*, 220, L15-L19. [2] Anders E. and Grevesse N. (1989) *GCA*, 53, 197-214. [3] Harper C. L. Jr. (1993) *J. Phys. G.*, 19, S81-S94. [4] Hidaka H. et al. (2000) *MAPS*, 35, A72-A73. [5] Hidaka H. et al. (2001) *Goldschmidt, Abstract #3318*. [6] Podosek et al. (1997) *MAPS*, 32, 617-627. [7] Podosek et al. (2000) *GCA*, 63, 2351-2362. [8] Podosek et al. (2000) *GCA*, submitted. [9] Zinner E. K. (1997) *AIP Conf. Proc.* 402, 3-26. [10] Nichols R. H. Jr. (1992) *LPSC XXIX*, 987-988.