assumption of uniform aerosol distribution; the main advantage is that the optical property (τ) of most interest to atmospheric scientists is directly indicated. This technique has been applied to 21 eclipses to yield a comparison of four (possibly five) volcanic aerosol events between 1960 and 1982. It may be possible to extend the τ_z time series back to 1600 A.D. (*27*). A long time series of global aerosol loading could be compared with time series based on historic and geologic eruption records and ice cores.

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19. Many observers classify the appearance of lunar eclipses according to a five-category luminosity scale devised by A. Danjon [see (*6*), p. 97]. For the six eclipses for which direct brightness observations are not available, published luminosity values have been converted into visual magnitudes by an empirical relation derived from the other 15 eclipses.
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28. I thank Dr. Uwe Radok for his comments and encouragement.

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Barium Isotopes in Allende Meteorite: Evidence Against an Extinct Superheavy Element

Abstract. *Carbon and chromite fractions from the Allende meteorite that contain isotopically anomalous xenon-131 to xenon-136 (carbonaceous chondrite fission or CCF xenon) at up to 5 × 10^{11} atoms per gram show no detectable isotopic anomalies in barium-130 to barium-138. This rules out the possibility that the CCF xenon was formed by in situ fission of an extinct superheavy element. Apparently the CCF xenon and its carbonaceous carrier are relics from stellar nucleosynthesis.*

Primitive meteorites contain a peculiar xenon component that is enriched up to twofold in the heavy isotopes ^{131}Xe to ^{136}Xe (*1*). The isotopic pattern resembles that of xenon from fission of actinides, and so this component has become known as CCF (carbonaceous chondrite fission) xenon. A possible source is an extinct superheavy element from the hypothetical "island of stability" (*2*) centered on the doubly magic nucleus ^{298}114 (*3*). An alternative possibility is direct nucleosynthesis of the heavy xenon isotopes, by neutron capture (*r*-process) in a supernova (*4, 5*).

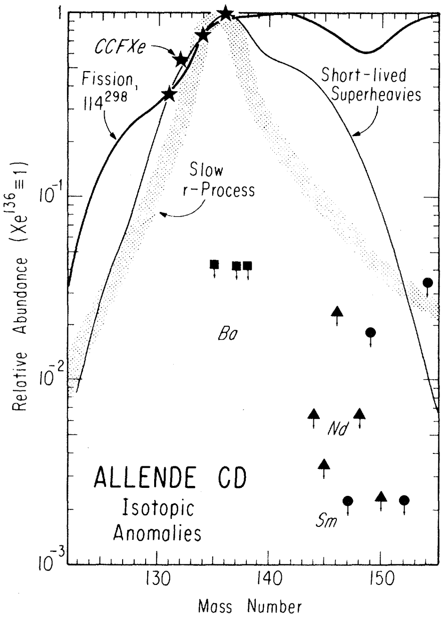

Fig. 1. Etch fractions from Allende CD that are highly enriched in CCFXe show no detectable anomalies for the neighboring elements Ba, Nd, and Sm. Upper limits range from 0.002 to 0.03 times the number of anomalous ^{136}Xe atoms, much less than predicted by the two principal models: in situ fission of a long-lived superheavy element (*7*) such as ^{298}114 (*17*), or neutron capture in stars (*4*), yielding nuclides near mass 140 either by direct buildup in a slow *r*-process (*5*) or by fission of short-lived superheavy elements of mass near 280 (*17*). Any process that made xenon isotopes 134 and 136 should have made comparable amounts of barium-135 to barium-138. The neutron capture model can escape this predicament by assuming that the Ba, Nd, and Sm expelled from the star condensed in the first crop of dust grains (oxides), whereas Xe condensed in a later crop (carbon). No such excuse is available for the in situ fission model, which is therefore ruled out by these data.

Until now, these two hypotheses have remained in a virtual stalemate (*6*). The fission model is supported by the correlation of CCFXe with volatile elements (*7, 8*), but it must invoke rather contrived explanations for the localization of CCFXe in a minor (~ 0.05 percent) carbon fraction in the meteorite (*8, 9*), and for its close coupling with another anomalous xenon component (L-Xe), enriched in isotopes 124 to 130 (*4*). The nucleosynthesis model can, in principle, account for these two trends, but has trouble explaining why the carbonaceous carrier of CCFXe is isotopically normal (*6, 9*), or why CCFXe is not accompanied by large amounts of ^{129}Xe from the decay of ^{129}I, of half-life 15 × 10^6 years (*10*).

An obvious test of both models had become feasible since the discovery of the carrier of CCFXe (*8*): detection of isotopic anomalies in neighboring elements, especially Ba and the light rare earth elements (REE) (*11*). With additional data points in the mass range 130 to 154, the isotopic pattern may be sufficiently well defined for a conclusive match or mismatch. However, the expected anomalies are on the order of 10^{11} atoms per gram of carrier, and hence require samples of very low Ba and REE content as well as careful chemistry and mass spectrometry. An early attempt gave negative but inconclusive results (*11*).

We prepared a sample strongly enriched in CCFXe by dissolving a large piece of the Allende C3V chondrite in HF-HCl, and removing noncolloidal material—mainly spinel—from the carbonaceous, acid-insoluble residue (*12*). We then treated this sample (Allende CD) with successively stronger oxidants to dissolve its main phases: (i) Q, the carrier of the dominant, normal xenon component (probably not a true phase, but merely an adsorbed surface layer); (ii) Cγ, gas-poor carbon; (iii) Cδ, gas-rich, resistant carbon, containing most of the CCFXe, and (iv) chromite (*9*) (Table 1). Barium, neodymium, samarium, and strontium were measured in the solu-

Table 1. Stepped oxidation of samples (all values for solutions are italicized).

Sample or *solution*	Weight (%)	Treatment (acid, °C, hours)	Phases* dissolved	^{132}Xe 10^{-8} (cm^3/g)	^{136}Xe/^{132}Xe	Concentration (10^{11} atoms per gram of CD)	
						^{136}Xe†	^{135}Ba†
CD	≡ 100‡	HNO_3, 75, 2		40.4	0.357	5.13	
CE-1			*Q*			*0.88*	*< 0.036*
CE	97.8	$HClO_4$, 140, 2		11.6	0.449	4.25	
CF-1			*C*γ			*0.20*	*< 0.14*
CF	28.4	$HClO_4$, 190, 2					
CG-1			*C*δ, *Chr*			*(2.95)*	*< 0.21*
CG	9.8			12.3	0.655	1.10	

*Q, carrier of isotopically normal, "planetary" xenon; $C\gamma$, reactive carbon, containing little or no anomalous xenon; $C\delta$, less reactive carbon, containing CCFXe; *Chr*, chromite. †Isotopically anomalous components. The ^{136}Xe values are calculated relative to normal xenon of $^{136}Xe/^{132}Xe \equiv 0.310$. The values for solutions are calculated by difference (*14*). The ^{135}Ba values are upper limits, based on the standard normalization. ‡This sample, weighing 152 mg, represents 0.223 percent of the bulk meteorite.

tions (*13*), whereas xenon was measured in residues (*14*); xenon values for solutions were calculated by difference.

The isotopic data for Ba are shown in Table 2, those for Nd, Sm, and Sr are given in the following report (*15*). Clearly, there are no anomalies whatsoever. For our purposes, ^{135}Ba is especially important, as it lies between two of the isotopes of CCFXe, ^{134}Xe and ^{136}Xe. We therefore calculated upper limits to any anomalous ^{135}Ba (*16*) for comparison with anomalous ^{136}Xe in the same fraction (Table 1).

Evidently the upper limits on a ^{135}Ba anomaly are only 0.04 to 0.07 times as great as the ^{136}Xe anomalies in the same fractions. This is a major embarrassment for all models—fission or nucleosynthesis—as any process that makes ^{134,136}Xe must make comparable amounts of ^{135}Ba. Given the volatility of Xe, one would expect nonvolatile elements to have larger, not smaller, absolute anomalies.

The magnitude of the problem is illustrated in Fig. 1, showing the predictions of the fission and nucleosynthesis models together with upper limits for possible Ba, Nd, and Sm anomalies. (To avoid clutter, we have plotted only the lowest of the 3 to 11 values for each isotope.) Three predicted patterns are shown: fission of the long-lived doubly magic nuclide $^{298}114$ (*17*); neutron capture [similar to the *r*-process, but at lower neutron densities, $\sim 10^{20}$ versus $\sim 10^{28}$ cm^{-3} (*5*, *18*)]; and fission of a mix of short-lived superheavy nuclides of mass around 280 made by some neutron capture process (*17*, *18*).

None of the three patterns predicts such small anomalies for the unshielded Ba, Nd, and Sm isotopes. We can at once dismiss the trivial explanation that anomalous Ba, Nd, and Sm were leached out of Allende CD during the month-long treatment with HCl and HF, or that they exchanged with blank Ba, Nd, and Sm in the acids. Not only have CCFXe and two large Nd anomalies survived this treatment (*15*), but—especially to the point—so has highly radiogenic Sr (*15*), the chemical congener of Ba. Thus, it is very unlikely that other anomalies for Ba, Nd, and Sm were depleted by two orders of magnitude during this treatment.

For the nucleosynthesis models, chemistry offers a way out of this dilemma. Barium, neodymium, and samarium all have high condensation temperatures but low first ionization potentials compared to Xe. Accordingly, these three elements may have been separated from Xe by prior condensation on an earlier crop of dust grains in the expanding supernova shell or by plasma processes (*19*).

Table 2. Isotopic composition of barium in Allende samples (ϵ^i = deviation of ^{i}Ba/^{138}Ba from normal Ba, in parts per 10^4). Values are corrected for mass discrimination to $^{135}Ba/^{138}Ba = 0.091940$. The normal ratios, ^iR, relative to ^{138}Ba, are: $^{137}R = 0.156545 \pm 3$; $^{136}R = 0.109543 \pm 4$; $^{134}R = 0.033716 \pm 3$; $^{132}R = 0.001413 \pm 1$; $^{130}R = 0.001475 \pm 1$.

ϵ^{137}	ϵ^{136}	ϵ^{134}	ϵ^{132}	ϵ^{130}	Ba (ppb)*
		CE-1			
−1.0 ± 1.0	+0.5 ± 1.3	+0.6 ± 2.4	+5.7 ± 21.2	+11.5 ± 27.1	65
		CF-1			
−0.3 ± 0.3	+0.5 ± 0.5	+1.2 ± 1.2	−1.4 ± 14.2	+4.7 ± 13.6	830
		CG-1			
−0.5 ± 0.9	+0.5 ± 1.3	+1.2 ± 1.5	+17.0 ± 15.5	−2.0 ± 13.6	800
		Tholeiite E-2a (10 ng)			
−0.3 ± 0.3	−0.3 ± 0.5	−0.3 ± 1.2	+2.8 ± 7.1	+11.5 ± 6.8	

*Relative to parent sample, Allende CD.

No such excuse is available for the in situ fission model, as all fission products ought to be trapped in the carbon carrier with nearly equal efficiency. Thus the present data rule out a long-lived, fissioning element as the source of CCFXe, leaving nucleosynthesis as the only alternative. A direct argument for nucleosynthesis is the recent discovery of anomalous nitrogen, depleted in ^{15}N by up to 33 percent, in association with CCFXe (*20*). Thus the stalemate over the origin of CCFXe has at last been broken, 18 years after its discovery. Apparently CCFXe and its carbonaceous carrier represent relict matter from stellar nucleosynthesis that somehow survived the formation of the solar system.

The absence of a ^{135}Ba excess allows us to place a tentative upper limit on the abundance of its radioactive progenitor, ^{135}Cs (half-life = 3×10^6 years). A similar, though less purified Allende residue, 3C1, contained 7×10^{14} atoms per gram of stable ^{133}Cs (*7*); if we assume that Allende CD had the same Cs concentration, then its initial $^{135}Cs/^{133}Cs$ ratio was 6×10^{-5}. This is low compared to the steady-state ratio for galactic nucleosynthesis, 6×10^{-4} (*21*), and suggests a decay interval of $> 10^7$ years between cessation of nucleosynthesis and formation of the Cs-bearing phases in the Allende residue.

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remove Cr from solutions CF-1 and CG-1, gaseous HCl was bubbled through the $HClO_4$ solution for 20 minutes at 180°C. Ba, Nd, Sm, and Sr were separated from unspiked and spiked (10 percent) aliquots of the etch solutions, using ion exchange procedures; the spike contained enriched isotopes of the first three elements but not Sr. Mass spectrometric procedures were developed that yield precise data for major isotopes for samples as small as 1 ng. Normal isotopic ratios were determined from a standard solution made from high purity $BaCO_3$. As a control sample, a terrestrial tholeiite (E-2a) from eastern Egypt was used.

14. As no aliquot of CF had been retained, we apportioned the gas loss from CE to CG according to that for an analogous set of samples, prepared from a different colloidal aliquot [Allende BB; table 3 of (9)] of the same acid-insoluble residue.
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22. We thank J. W. Morgan for illuminating discussions of the possible role of fission recoil. This work was supported in part by NASA grants NAG 9-52 and NAG 9-49. This is SIO-IG^2L contribution 13.

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Samarium-146 in the Early Solar System: Evidence from Neodymium in the Allende Meteorite

Abstract. *A carbon-chromite fraction from the Allende C3V chondrite shows strikingly large isotopic enrichments of neodymium-142 (0.47 percent) and neodymium-143 (36 percent). Both apparently formed by alpha decay of samarium-146 and samarium-147 (half-lives 1.03 × 10^8 and 1.06 × 10^{11} years), but the isotopic enrichment was greatly magnified by recoil of residual nuclei into a carbon film surrounding the samarium-bearing grains. These data provide an improved estimate of the original abundance of extinct samarium-146 in the early solar system [$^{146}Sm/^{144}Sm = (4.5 \pm 0.5) \times 10^{-3}$], higher than predicted by some models of p-process nucleosynthesis. It may be possible to use this isotopic pair as a chronometer of the early solar system.*

In an effort to understand an isotopically anomalous xenon component (CCFXe) in meteorites (1), we have looked for similar anomalies in the neighboring elements Ba, Nd, and Sm, as well as Sr. Most of our results were negative, as described in the preceding report (2), ruling out the possibility that the xenon component formed by in situ fission of an extinct superheavy element (3) and thus leaving stellar nucleosynthesis as the only alternative. However, we found unexpected anomalies in the neodymium isotopes 142, 143, 145, and 146, as well as a large enrichment in strontium-87. These will be discussed in the present report.

The starting material was a colloidal carbon plus chromite fraction (Allende CD) prepared by dissolving a sample of the Allende C3V chondrite in HF-HCl, and treating the insoluble residue (0.56 percent) to remove noncolloidal material. A 152-mg portion of Allende CD was then etched with successively stronger oxidants for 2 hours each, yielding three solutions (the mass fraction dissolved is given in parentheses): (i) CE-1: HNO_3, 70°C (2.2 percent); (ii) CF-1: $HClO_4$, 140°C (69.4 percent); and (iii) CG-1: $HClO_4$, 190°C (18.6 percent). Each solution was processed further by cation exchange to separate Sr, Ba, Nd, and Sm (2, 4).

Table 1. Strontium in etch fractions from Allende CD. Errors in this and the subsequent tables are at the 95 percent confidence level. Data were corrected for fractionation to $^{86}Sr/^{88}Sr = 0.1194$. For NBS standard 987, this yields $^{87}Sr/^{86}Sr = 0.710260$, with a total range of ± 0.000026.

Fraction	Phase*	$\frac{^{84}Sr}{^{88}Sr}$	$\frac{^{87}Sr}{^{86}Sr}$
CE-1 HNO_3 70°C	Surface (2.2)	0.006744 ± 10	1.24650 ± 10
CF-1 $HClO_4$ 140°C	Cγ (69)	0.006744 ± 8	0.72611 ± 7
CG-1 $HClO_4$ 190°C	Cδ (19)	0.006745 ± 6	0.70046 ± 6
Moore County plagioclase		0.006746 ± 3	0.699085 ± 25

*Phases dissolved; their mass fraction (percent) in the sample is shown in parentheses. Cγ, reactive carbon, of low noble-gas content. Cδ, resistant carbon, containing isotopically anomalous xenon (CCFXe) (2); this fraction also contains chromite.

Strontium. The three etch fractions show normal $^{84}Sr/^{88}Sr$ but high and variable $^{87}Sr/^{86}Sr$ (Table 1). This monoisotopic enrichment of ^{87}Sr apparently comes from radioactive decay of ^{87}Rb (half-life 4.88 × 10^{10} years). The extreme value for the HNO_3 fraction CE-1 implies a Rb/Sr ratio about ten times the solar system value if the sample is 4.56 × 10^9 years old. We cannot derive age information from the Rb and Sr data, however, as we did not measure the concentration of these elements (4).

Samarium and neodymium. Because of the very small sample weights (mostly 0.2 to 1 ng), the precison obtained was up to tenfold lower than usual. Like Ba, Sm shows no detectable isotopic anomalies (Table 2). In contrast, Nd shows large anomalies at masses 142 (up to 0.47 percent) and 143 (up to 36 percent), as well as small anomalies at 145 and 146 (Table 3). There is no doubt about the reality of the larger anomalies; they first showed up in preliminary experiments [^{143}Nd enriched by up to 6 percent, $^{87}Sr/^{86}Sr \simeq 0.822$ (5)], and then at nearly the present magnitude in ~ 10 percent portions of the present sample (6, 7). But before turning to the large anomalies at masses 142 and 143, let us examine the reality of the small anomalies at 145 and 146.

Neodymium-145 and -146. In our preliminary report (6) small splits from fractions CE-1 and CF-1 were measured on an electron multiplier (Daly detector); a larger split from CG-1 (which also contains more neodymium) was measured on the Faraday cup (Table 3). In the present work, the remaining, larger portions were measured on both the multiplier and the Faraday cup (Table 3). In order to check for nonlinearity of the multiplier system, a 1-ng sample of the La Jolla Nd standard was also measured on both detector systems (Table 3). These data show significant differences (bias) only at masses 143 and 145, with the electron multiplier values about 0.4 to 0.5 per mil low in both cases. The bias at 143 is inconsequential, in view of the large size of the meteoritic anomaly but the bias at 145 (−0.5 per mil) is significant. It could account for most of the anomaly in CE-1 and perhaps CG-1, but not in CF-1. Apparently CF-1 has significant anomalies at ^{145}Nd (−1 per mil) and ^{146}Nd (+1 per mil) (8), whereas CE-1 has an anomaly at ^{146}Nd.

These two nuclides are made by both the *s*- and *r*-processes, with the former estimated to contribute 25 and 45 percent, respectively, in normal solar system matter (9). Presumably sample CF-1