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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16. The data in Table 2 were all normalized to an assumed $^{135}Ba/^{138}Ba$ ratio of 0.091940, to correct for mass discrimination in the mass spectrometer. [This is the ratio normally used by us and others (O. Eugster, F. Tera, G. J. Wasserburg, *J. Geophys. Res.* **74**, 3897 [1969]) for this purpose]. Any ^{135}Ba excess would thus cause apparent deficiencies in all other Ba isotopes. Our estimates assume that no deficits are allowed on any of the isotopes; in addition to 2σ errors on the isotopic ratios, they also take into account blank contamination from the analytical procedures. These were highest for CE-1 ($\simeq$ 15 percent). We also calculated a second set of upper limits based on renormalization to $^{134}Ba/^{136}Ba$—the two major isotopes of Ba that are shielded from β^- decay. These limits are higher (< 0.18, < 0.21, and $< 0.42 \times 10^{11}$ atoms per gram of CD), especially for CE-1, but that merely reflects the larger error in the normalizing ratio caused by the low abundance of ^{134}Ba. We believe that the first set of upper limits is more realistic.
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21. For this calculation, we assume an age of 10^{10} years for the galaxy, an *r*-process production ratio P_{135}/P_{133} of 0.82, and an *s*-process contribution of 10 percent for ^{133}Cs (H. D. Zeh, private communication).
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-IG²L 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 $\pm$ 0.000026.

Fraction	Phase*	$^{84}Sr/^{88}Sr$	$^{87}Sr/^{86}Sr$
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

Table 2. Samarium in etch fractions from Allende CD. Results are given as per mil deviations from normal samarium: $\delta_i = 1000\,[({}^iR_{\text{sample}}/{}^iR_{\text{normal}}) - 1]$, where ${}^iR = {}^iSm/{}^{152}Sm$. All data were measured with an electron multiplier and were corrected for fractionation to ${}^{147}R = 0.56081$. The normal ratios are ${}^{154}R = 0.85078 \pm 6$, ${}^{150}R = 0.275974 \pm 28$, ${}^{149}R = 0.516826 \pm 16$, ${}^{148}R = 0.420469 \pm 51$, and ${}^{144}R = 0.114965 \pm 24$.

Sample	Sm (ng)	δ_{144}	δ_{148}	δ_{149}	δ_{150}	δ_{154}*	[Sm]†
CE-1	0.18	0.5 ± 6.2	−0.3 ± 1.5	+0.8 ± 1.1	+0.4 ± 2.2	+2.5 ± 6.4	0.53
CF-1	0.32	0.0 ± 2.8	+0.7 ± 0.9	−0.1 ± 1.2	+1.0 ± 1.4	+1.8 ± 3.3	0.93
CG-1	0.69	+0.5 ± 8.3	−0.5 ± 0.9	+1.0 ± 1.0	−0.4 ± 0.8	+1.4 ± 0.5	2.0

*Interference from ${}^{135}BaF^+$ (and ${}^{138}Ba^{16}O^+$?) not corrected. †10^{13} atoms per gram of Allende CD.

contains a Nd component with anomalous proportions of *s*- and *r*-process material. Such a component should have comparable anomalies at other isotopes as well, including 144 and possibly 148, the pair used for the mass discrimination correction, and so an accurate resolution of this component requires an elaborate renormalization and reanalysis of the data, for Nd as well as Sm and Ba. Such an analysis would be premature with the present data, as renormalization raises the errors, making the smaller anomalies quite marginal (*10*). We turn instead to ${}^{142,143}Nd$, whose anomalies are large enough to be essentially unaffected by the above ambiguities.

Neodymium-142 and neodymium-143. The most obvious source of the excess ${}^{142,143}Nd$ (henceforth denoted by asterisks) is α-decay of ${}^{146}Sm$ and ${}^{147}Sm$. However, there are two serious paradoxes. (i) With the largest observed ${}^{147}Sm/{}^{144}Nd$ ratio, 0.157 for CF-1, only 3.2 percent of the observed ${}^{143}Nd^*$ (relative to an initial ${}^{143}Nd/{}^{144}Nd$ of 0.50671) could form in 4.56×10^9 years. (ii) The ${}^{142}Nd^*$ requires an initial ${}^{146}Sm/{}^{144}Sm$ of 0.14, which seems much too high for an extinct radionuclide of this half-life, according to conventional nucleosynthesis models (*11*) and estimates of production ratios (*12, 13*).

Most alternatives we have considered do not work. Accelerated α-decay, at high temperatures in a stellar interior, could produce additional ${}^{143}Nd$, but only at the expense of a matching ${}^{147}Sm$ deficit, which is not observed. Photonuclear reactions at high temperatures (2.5×10^9 K) would build up rare earth element (REE) nuclides with 82 neutrons, including ${}^{143}Pm$, which would later decay to ${}^{143}Nd$. But they would also build up ${}^{144}Sm$, contrary to observation, while eroding ${}^{146}Sm$ (*11*).

The only satisfactory explanation we have been able to think of is recoil following α-decay of ${}^{147}Sm$ and ${}^{146}Sm$, which can propel the residual nucleus a few hundred angstroms through the crystal. (This mechanism is not available for the β-emitter ${}^{87}Rb$, as its recoil energy is some 3×10^{-5} times smaller.) Suppose a fraction of the Sm-bearing grains have carbon coatings, such as those observed for olivine and other minerals (*14*). Of the decays within recoil range of the surface (5 to 7 μg/cm^2), one-fourth will eject the residual ${}^{142,143}Nd^*$ nuclei into the carbon film. The ${}^{143}Nd^*$ in Allende CD represents 7.6×10^9 atoms per gram of meteorite, or some 1.3×10^{-3} of the total radiogenic ${}^{143}Nd$. In terms of the recoil model, such a fraction of trapped recoils would result if all the Sm were located in 10-μm grains, or 1 percent in 0.1-μm grains, and so on, all with the same average Sm concentration as the bulk meteorite. Many other combinations of grain size and Sm content are possible, of course.

Samarium-146. The above recoil mechanism greatly magnifies anomalies resulting from α-decay, by transferring the decay products to a phase of low REE content. This is particularly helpful for ${}^{142}Nd$, the decay product of ${}^{146}Sm$, as previous evidence for this nuclide, in the Angra dos Reis (AdoR) meteorite, has been marginal (*15*) and remained marginal upon reanalysis in another laboratory (*16*). In the present sample, ${}^{142}Nd$ is unmistakably enriched, by up to 50 standard deviations (Table 3), thus permitting an accurate estimate of the original ${}^{146}Sm$ abundance. If recoil indeed is the cause of the ${}^{142,143}Nd$ enrichments, then the ratio ${}^{142}Nd^*/{}^{143}Nd^*$ reflects ${}^{146}Sm/{}^{147}Sm$ at the time the carbon layer was deposited on the grains. If this event occurred $\sim 4.56 \times 10^9$ years ago, then the mean ${}^{146}Sm/{}^{147}Sm$ ratio at that time was $(0.93 \pm 0.10) \times 10^{-3}$ (Table 4) (*17*). This value agrees quite well with the earlier result for the Ca-rich achondrite AdoR $(0.96 \pm 0.47) \times 10^{-3}$ (*15*), but the uncertainty now is only 11 percent.

A perennial question that must be asked in each such case is whether the decay of the extinct radionuclide took place in the early solar system or in a more ancient, interstellar or circumstellar, environment (*18*). For AdoR, a local origin is certain: this highly differentiated meteorite can have formed only by igneous processes in a planetoidal body, not in interstellar space. Moreover, this meteorite is firmly linked to the solar system by its age of 4.56×10^9 years, as determined by several different radiometric dating methods (*15, 19*). For

Table 3. Neodymium in etch fractions from Allende CD. Results are given as per mil deviations from normal neodymium: $\delta_i = 1000\,[({}^iR_{\text{sample}}/{}^iR_{\text{normal}}) - 1]$, where ${}^iR = {}^iNd/{}^{144}Nd$. All data were measured as NdO^+ and were corrected for fractionation to ${}^{148}R = 0.241572$. Normal Nd (La Jolla standard) is: ${}^{142}R = 1.141826 \pm 11$, ${}^{143}R = 0.511858 \pm 4$, ${}^{145}R = 0.348419 \pm 2$, ${}^{146}R = 0.721878 \pm 7$, and ${}^{150}R = 0.236428 \pm 7$.

Sample	Nd (ng)	Detector*	δ_{142}	δ_{143}†	δ_{145}	δ_{146}	δ_{150}	[Nd]‡
CE-1	1.1	M§	3.9 ± 0.8	335.3 ± 0.5	−0.7 ± 0.4	1.3 ± 0.4	0.5 ± 1.0	3.6
		M	4.41 ± 0.22	362.60 ± 0.22	−0.86 ± 0.11	1.05 ± 0.15	0.4 ± 0.5	
		F	4.47 ± 0.24	364.03 ± 0.27	0.09 ± 0.31	0.88 ± 0.21	0.2 ± 0.5	
CF-1	1.2	M§	4.0 ± 0.5	271.4 ± 0.5	−1.9 ± 0.6	1.2 ± 0.5	0.1 ± 0.7	3.9
		M	4.47 ± 0.19	277.15 ± 0.25	−1.58 ± 0.29	1.05 ± 0.16	0.5 ± 0.5	
		F	4.69 ± 0.18	277.59 ± 0.14	−1.04 ± 0.26	1.04 ± 0.21	−0.3 ± 0.4	
		F	4.79 ± 0.24	277.89 ± 0.17	−1.09 ± 0.17	1.05 ± 0.14	−0.7 ± 0.3	
CG-1	3.0	F§	1.3 ± 0.2	63.4 ± 0.2	−0.2 ± 0.3	0.2 ± 0.2	−0.3 ± 0.5	9.3
		M	1.30 ± 0.22	63.14 ± 0.23	−0.54 ± 0.30	0.19 ± 0.19	0.4 ± 0.5	
		F	1.49 ± 0.13	63.67 ± 0.11	−0.39 ± 0.30	0.10 ± 0.13	−0.4 ± 0.3	
		F	1.39 ± 0.13	63.62 ± 0.14	−0.23 ± 0.19	0.16 ± 0.16	−0.1 ± 0.4	
Standard	1.0	M	−0.04 ± 0.18	−0.38 ± 0.19	−0.49 ± 0.17	0.08 ± 0.12	0.1 ± 0.2	
		F	0.06 ± 0.16	0.05 ± 0.14	−0.02 ± 0.15	−0.03 ± 0.07	0.1 ± 0.4	

*M, electron multiplier; F, Faraday cup. †δ_{143} for the Allende fractions was calculated relative to ${}^{143}R$ for the Juvinas meteorite ≡ 0.512566. ‡10^{13} atoms per gram of Allende CD. §Values in this row are from (*6*).

Allende, an undifferentiated meteorite, a fossil, presolar contribution is more plausible, especially as the evidence for ^{146}Sm is based on only 1.3×10^{-3} of the total radiogenic ^{142,143}Nd. However, the close agreement with the indisputably local AdoR speaks for a common, local source, as does the absence of significant anomalies in the nonradiogenic isotopes of Nd, Sm, Sr, and Ba in CE-1. Very probably both the Sm-bearing grains in Allende and their carbon coatings formed in the early solar system.

Implications for nucleosynthesis. Samarium-146, being shielded from β^--decay by ^{146}Nd, can form only by the *p*-process (proton capture or photodisintegration), and hence can provide some insights into this nucleosynthesis process. For this purpose, we express its abundance relative to that of ^{144}Sm, another pure *p*-process isotope: $^{146}\text{Sm}/^{144}\text{Sm} = (4.5 \pm 0.5) \times 10^{-3}$ (Table 4). As Sm and Nd in Allende CD (and AdoR) have normal solar system isotopic composition within $\simeq$ 2 per mil, the above ratio must be very close to the average *p*-process contribution for the solar system. We can estimate from that ratio the production ratio of these two isotopes by the equation of Schramm and Wasserburg (*20*), on the assumption (*13*) that they were made by continuous nucleosynthesis for at least a few hundred million years, followed by a free decay interval of, say, $\sim 150 \times 10^6$ years between isolation of solar system material and formation of the meteorites. We obtain a production ratio $P_{146}/P_{144} = 0.42$, close to estimates of Audouze and Schramm (0.35 to 0.60) (*12*) and Lugmair *et al.* (0.57) (*13*) but much higher than that of Woosley and Howard (~ 0.02) (*11*). The latter estimate was based on the large (γ,n) cross section of ^{146}Sm and a partial bypass by the photodisintegration flow of ^{146}Gd, a progenitor of ^{146}Sm. Our value suggests that some of the assumptions of (*11*) may have to be revised. The average photodisintegration flow in this mass range may, in fact, have occurred at higher neutron numbers (that is, lower temperatures), and the (γ,α) reaction on ^{150}Gd, augmented by later α-decay of residual ^{150}Gd, may have made substantial contributions to ^{146}Sm.

A further possibility for the production of *p*-nuclei (*21*) is that "seed" nuclei were exposed to a thermal radiation field at $kT \sim 550$ keV, where k is the Boltzmann constant and T is temperature. Because (γ,n) reaction rates are slightly higher than thermal emission rates of particles, the disintegration path would be shifted to the proton-rich side of the β-stability valley for times as short as 10^{-8} second, before β^+-decay can occur. The mass number at which the disintegration path intercepts the closed 82-neutron shell depends critically on T. Consequently, for this process the $^{146}\text{Sm}/^{144}\text{Sm}$ ratio may serve as a sensitive temperature indicator for the production of neutron-poor nuclei above mass number $\simeq$ 110.

Whereas the AdoR data were only a strong hint for the existence of ^{146}Sm in the early solar system, the new results provide conclusive evidence. The abundance ratio of $^{146}\text{Sm}/^{144}\text{Sm}$, and the agreement of the inferred production ratio with two earlier estimates (*12, 13*), suggest that a "terminal spike" of nucleosynthesis could have made at most a very small contribution to the overall abundance of *p*-nuclei in solar system material. This is also true of a late-stage injection of supernova material, as such a contribution is estimated to have been only $\sim 10^{-4}$ (*22*).

Location of anomalies within carbon types. The various isotopic components show distinctive distributions among the three etch fractions. The radiogenic components (^{87}Sr, ^{142,143}Nd), of local origin, are contained mainly in the first two fractions (Cγ), whereas the nucleosynthetic CCFXe is contained mainly in the third fraction (Cδ), which also contains highly anomalous nitrogen (*23*). Yet this third fraction shows no comparable anomalies in Ba, Nd, Sm, and Sr. Presumably these elements became separated from C, N, and Xe in their presolar place of origin, by condensing on a separate crop of grains (*2*).

Table 4. Abundance of ^{146}Sm 4.56×10^9 years ago.

Fraction	$^{146}\text{Sm}/^{147}\text{Sm} \times 10^3$*	$^{146}\text{Sm}/^{144}\text{Sm} \times 10^3$†	Reference
CE-1	0.70 ± 0.18	3.4 ± 0.9	(*6*)
	0.73 ± 0.07	3.6 ± 0.3	
	0.72 ± 0.07	3.5 ± 0.3	
Weighted mean	*0.72 ± 0.02*	*3.5 ± 0.1*	
CF-1	0.88 ± 0.15	4.3 ± 0.7	(*6*)
	0.96 ± 0.09	4.7 ± 0.4	
	0.99 ± 0.09	4.8 ± 0.4	
	1.01 ± 0.10	4.9 ± 0.5	
Weighted mean	*0.97 ± 0.06*	*4.7 ± 0.3*	
CG-1	1.19 ± 0.23	5.8 ± 1.1	(*6*)
	1.29 ± 0.25	6.3 ± 1.2	
	1.37 ± 0.17	6.7 ± 0.8	
	1.27 ± 0.17	6.2 ± 0.8	
Weighted mean	*1.29 ± 0.11*	*6.3 ± 0.5*	
Grand avg.‡	0.93 ± 0.10	4.5 ± 0.5	
AdoR	0.96 ± 0.47	4.7 ± 2.3	(*15*)

*Based on $\delta^{142}\text{Nd}/\delta^{143}\text{Nd}$ from Table 3, with an 11 percent correction for the greater recoil energy of ^{142}Nd, and a small correction for bias of the electron multiplier. †For normal $^{144}\text{Sm}/^{147}\text{Sm}$, from Table 2. ‡Mass-weighted, by the mass of excess ^{143}Nd.

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7. The somewhat smaller excesses on ^{142,143}Nd in the earlier data (Table 3) are due to dilution with blank neodymium, which affected the main fractions (~ 90 percent) to a much lesser degree. Some blank neodymium may also have adsorbed or exchanged during the initial HCl-HF treatment, but that would only lower the excesses of ^{142,143}Nd without altering their ratio.
8. Although we use ^{146}Nd spike in the laboratory, we consider it very unlikely that the ^{146}Nd anomaly actually represents spike contamination. Not only would such contamination have to occur for all three samples, each time scaled to the amount of Nd in the sample so as to give the same relative anomaly, but it also would be accompanied by ^{149}Sm contamination, as we use a single spike solution containing both isotopes.
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from the relation $^{146}Sm/^{147}Sm \simeq \lambda_{147}t$ $(^{142}Nd/^{143}Nd)_{ch}(^{142}\delta/^{143}\delta)/1.11$, where t is the age of Allende CD (assumed 4.56×10^9 years), λ is the decay constant of ^{147}Sm, δ is defined in Table 2, ch is chondritic, and 1.11 is the ratio of the recoil ranges of ^{142}Nd and ^{143}Nd.
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24. We thank H.-G. Stosch for his invaluable help with the chemical separation of REE and Sr and N. Sugarman for discussion of recoil effects and accelerated decay. This work was supported in part by NASA grants NAG 9-49 and NAG 9-52. This paper is SIO-IG^{2}L contribution 14.

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El Chichón: Composition of Plume Gases and Particles

Abstract. *Aircraft measurements were made of trace gases, atmospheric particles, and condensed acid volatiles in the plume of El Chichón volcano, Chiapas, Mexico, in November 1982. Hydrogen sulfide was the primary gaseous sulfur species in the plume at the time of collection. Concentrations of 28 elements were determined by neutron activation analysis of particulate material from the plume. Rates of trace element emission to the atmosphere for each species were estimated by normalization to the simultaneously determined total sulfur emission rate. The volatile elements sulfur, chlorine, arsenic, selenium, bromine, antimony, iodine, tungsten, and mercury were enriched relative to bulk pyroclastic material by factors of 60 to 20,000. Arsenic, antimony, and selenium were associated predominantly with small (≤ 3 micrometer) particles. Calcium and sodium were present almost exclusively on larger particles and aluminum and manganese were bimodally distributed. Ash-laden particulate material injected into the stratosphere during the early violent eruptions was enriched by factors of 10 to 30 relative to ash in some of the same elements observed in the quiescent plume.*

Gases and particulate material emitted to the atmosphere by volcanic activity have been suggested as a major source of anomalously enriched elements in remote background aerosols (*1*). Aircraft studies (*2–4*) have confirmed ground studies (*5, 6*) showing that noneruptive volcanic outgassing produces aerosols highly enriched in volatile and chalcophilic elements.

The Mexican volcano El Chichón underwent violent eruption in late March and early April 1982, injecting large quantities of gas and ash above the tropopause, producing the largest stratospheric cloud of volcanic origin observed this century. Atmospheric turbidity and temperature anomalies have been reported (*7, 8*) and are still being monitored. We investigated the chemical nature of the material emitted that is most likely to exert long-range effects on the global background aerosol. This study of the emissions from El Chichón was pursued as a part of the Research on Atmospheric Volcanic Emissions (RAVE) project, a joint effort by the National Aeronautics and Space Administration in cooperation with five universities.

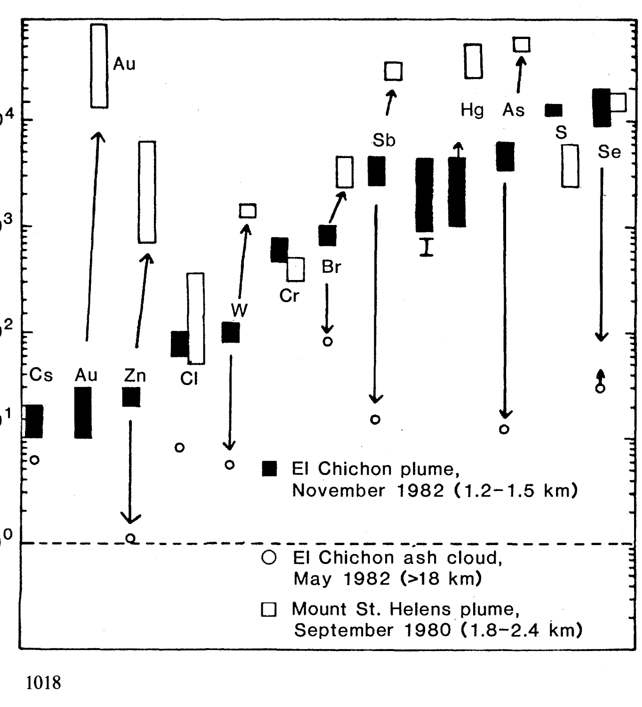

Fig. 1. Enrichment factors (relative to volcanic ash) for selected volatile elements on particles from the El Chichón plume.

All samples collected by us were obtained with NASA aircraft 429, a Lockheed Electra turboprop modified for tropospheric gas and particle collection (*9*). Continuous total gaseous sulfur measurement was provided by an onboard Meloy flame photometric detector (FPD) interfaced to a chromatographic system for the in-plume determination of H_2S and SO_2.

Samples were collected on 3 November 1982, at which time El Chichón was emitting a visible plume to an altitude of 0.7 km. The plume had a width of 3 km approximately 1 km downwind from the crater rim. Two sets of filter samples and one cascade impactor sample were collected while the plane flew coaxially in the plume. Teflon filters were selected for their high flow rate and low blank characteristics. Two types of multiple filter stacks were employed. One type contained a single Teflon filter followed by four cellulose filters that had been treated with 1*M* ^7LiOH in 30 percent glycerol to trap acid volatiles (such as SO_2, HF, HCl, and HBr). The second consisted of a quartz fiber filter followed in sequence by two nylon filters and two cellulose filters that had been treated with 1*M* $NaHSO_4$ in 10 percent glycerol for the collection of acidic gases (HCl and HNO_3) and ammonia, respectively. The cascade impactor collected size-segregated particles on polycarbonate films on seven stages followed by a polycarbonate backup filter. Blanks were collected for all filter and film types by exposing them for 30 seconds. Background air filter samples were collected while the plane flew at an altitude of 3 km to and from the volcano.

In addition to our own samples, we analyzed a sample of ash injected above the tropopause by the March and April 1982 eruptions of El Chichón that was collected by the NASA-DOE (Department of Energy) WB57F aircraft. This sample was collected above 18 km in May 1982 over the continental United States.

All filters were preloaded in cassette holders, stored in cleaned polyethylene bags before sample collection, and returned to clean bags immediately thereafter. Teflon and ^7LiOH-glycerol–treated cellulose filters, returned to the University of Maryland, were prepared in a class 100 clean room and tested for 28