

ANOMALOUS Ca ISOTOPIC COMPOSITIONS IN LEACHATES OF MURCHISON. J. H. Chen¹, D. A. Papanastassiou², and N. Dauphas³, ^{1,2}Science Division, ¹M/S 183-601, ²M/S 183-335, Jet Propulsion Laboratory, Caltech, 4800 Oak Grove Dr., Pasadena, CA 91109-8099 (James.H.Chen@jpl.nasa.gov), ³Origins Lab, Dept. Geophysical Sciences and Enrico Fermi Institute, University of Chicago, 5734 South Ellis Avenue, Chicago, IL 60637.

Introduction: Last year we presented Cr measurements on leachates from Murchison and on residues from stepwise acid dissolution of Orgueil and Murchison [1, 2, 3]. We identified large effects in bulk residues and leachates. We also measured *in situ* by SIMS the Cr isotopic composition of a residue from the Orgueil meteorite [3]. This work has identified the elusive carrier (or one of the carriers) of ⁵⁴Cr effects as nanospinel of supernova origin. Similar effects have also been independently presented by [4]. The recent Cr work has followed extensive searches by other groups [5, 6, 7]. The possible nucleosynthetic sources for the ⁵⁴Cr effects were reviewed by [3]. These workers pointed out that “measuring the isotopic abundance of ⁴⁸Ca in nanospinel will provide a definitive test to distinguish between SNIa and SNII origins, as high density SNIa represent the only conceivable stellar source for ⁴⁸Ca [8, 9].” Based on this expectation we have extended the techniques developed in our laboratories [10] to the measurement of ⁴⁶Ca and ⁴⁸Ca and applied them to the measurement of the Ca isotopic composition of leachates and residues from Murchison. The dissolution steps for samples in this work were described in [1, 3] and originally in [11], in connection with Os isotope measurements.

Analytical methods: We separated Ca from the leachate solutions using anion and cation exchange resins to eliminate other elements which might interfere with the measurement of Ca isotopes (K, Ti). Following the procedure of [10] we use the multidynamic mode for measuring Ca isotopes. The data are normalized for isotope fractionation to ⁴²Ca/⁴⁴Ca = 0.31221. It is recognized that measuring Ca isotopes to high precision is challenging, due to the low abundance of Ca isotopes other than ⁴⁰Ca and their large mass dispersion (20%). Based on repeat analyses of a terrestrial standard, we established the external precision (reproducibility between runs, at the 1 σ level of the distribution): ± 0.61 , ± 0.58 , ± 13.4 , and ± 0.9 ϵ u (parts in 10⁴) for ⁴⁰Ca, ⁴³Ca, ⁴⁶Ca and ⁴⁸Ca (plotted as error envelopes in Fig. 1). These are the levels of precision that permit the resolution of isotope anomalies for Ca in our work. In Table 1 and Figures we show the intrinsic precision ($2\sigma_{\text{mean}}$) for individual samples, as a measure of the quality of each run. The resolution of anomalies is based on the external precision estimates (unless the internal precision is worse, e. g., as for Step 7, due to small sample size).

Results and discussion: The Ca results for the Murchison samples are plotted in Figure 1. For all samples 1-3 μ g of highly purified Ca were loaded using double filament assemblies and analyzed. Since only a few hundred ng of Ca were obtained for Step 7, the precision of the data is worse. We monitored the Ti interferences by measuring the intensities at 47 and 49

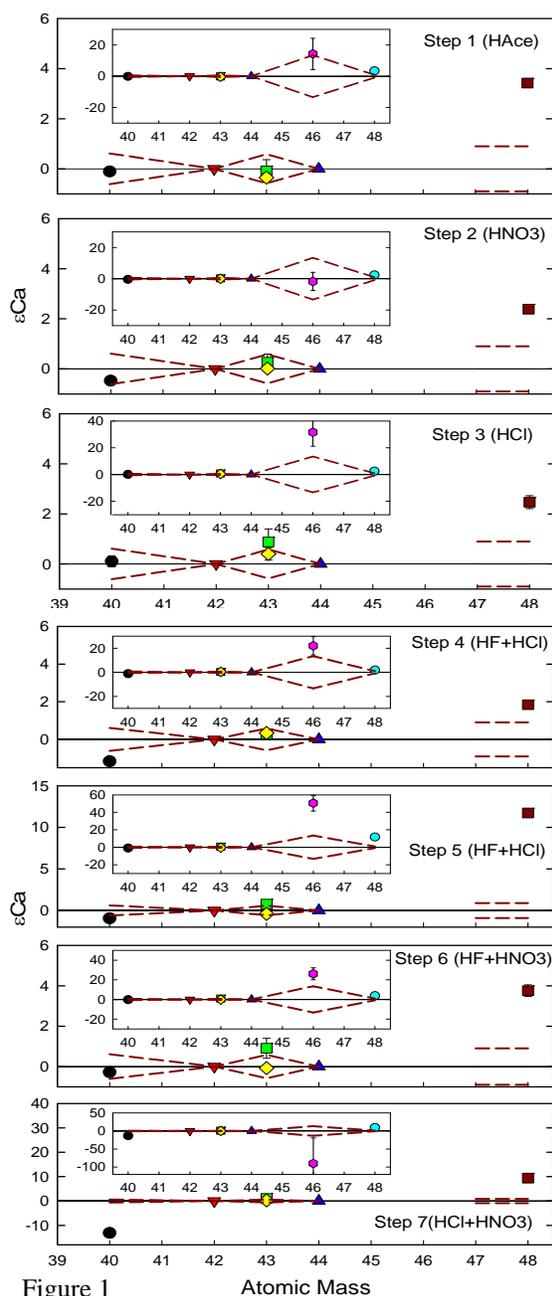
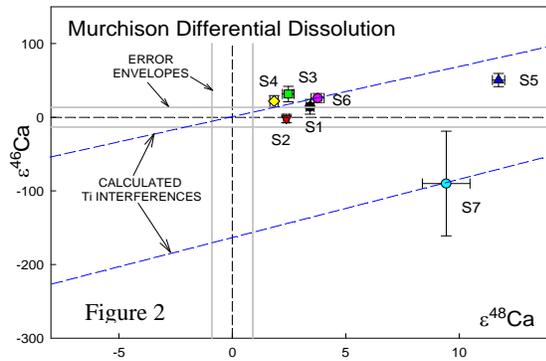


Figure 1

Atomic Mass

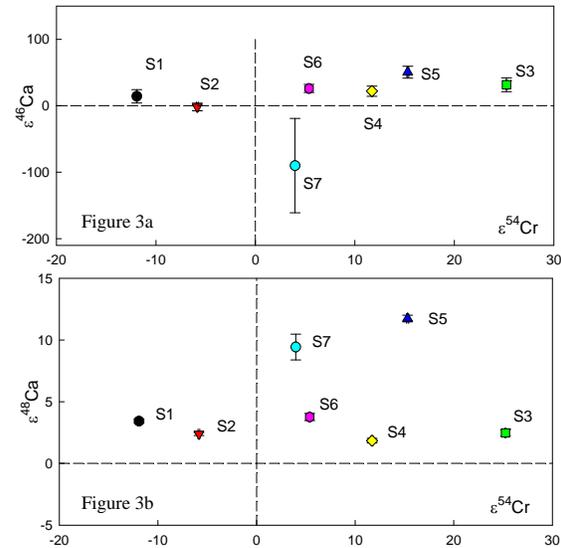


amu. For all samples, there were no resolvable signals (using the Faraday cup) above the base lines at these masses. Therefore, no correction from Ti interferences was needed. However, because of the very low ^{46}Ca abundance (30 ppm), the results on ^{46}Ca are very sensitive to any interference from Ti. For all Murchison samples there are no resolvable $\epsilon^{43}\text{Ca}$ effects. Most samples also show no effect in $\epsilon^{40}\text{Ca}$ (with a resolution of $\pm 0.61 \text{ \epsilon u}$) except small -1.2 ± 0.1 and $-0.9 \pm 0.2 \text{ \epsilon u}$ effects in Steps 4 and 5 and a large -13 \epsilon u in Step 7. For $\epsilon^{46}\text{Ca}$, even with the larger uncertainties, the data appear to show excesses for Steps 3-6 and a deficit for Step 7. All Murchison samples show resolvable excesses on $\epsilon^{48}\text{Ca}$ ranging from $+2$ to $+12 \text{ \epsilon u}$. In a $\epsilon^{46}\text{Ca}$ versus $\epsilon^{48}\text{Ca}$ diagram (Fig. 2), we show the effect of possible Ti interference on the Ca data. All Murchison samples except S2, S5 and S7 plot close to a mixing line between normal Ca and the Ti end member. However, the implied corrections for Ti interference are too large, relative to our measurements of $^{47,49}\text{Ti}$ interferences. Hence, we believe that these samples require at least a distinctly anomalous ^{48}Ca isotopic composition. In Fig. 3 we compare the $\epsilon^{54}\text{Cr}$ and the $\epsilon^{46}\text{Ca}$, $\epsilon^{48}\text{Ca}$ values for each sample. The data clearly show that there are definite ^{48}Ca and possible ^{46}Ca effects in

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Table 1. Ca isotopic composition in samples of Murchison

Name	$\epsilon^{40}\text{Ca}$	$\pm\text{error}$	$\epsilon^{43}\text{Ca-I}$	$\pm\text{error}$	$\epsilon^{43}\text{Ca-II}$	$\pm\text{error}$	$\epsilon^{46}\text{Ca}$	$\pm\text{error}$	$\epsilon^{48}\text{Ca}$	$\pm\text{error}$
S1	-0.11	0.13	-0.08	0.44	-0.35	0.18	14.27	10.03	3.43	0.17
S2	-0.47	0.12	0.26	0.33	0.01	0.12	-1.76	5.79	2.38	0.13
S3	0.10	0.22	0.88	0.52	0.41	0.25	31.59	10.47	2.47	0.26
S4	-1.16	0.10	0.28	0.21	0.34	0.15	21.97	7.61	1.84	0.19
S5	-0.94	0.19	0.76	0.45	-0.41	0.19	50.51	8.96	11.74	0.27
S6	-0.27	0.18	0.91	0.49	-0.07	0.12	26.05	6.12	3.76	0.29
S7	-13.10	0.48	1.04	1.41	0.25	1.17	-90.00	70.97	9.43	1.05



samples that show definite and large ^{54}Cr effects. This can be viewed to first order as a correlation. However, there is no specific correlation between the sizes of the ^{54}Cr and ^{48}Ca effects. Such a lack of proportionality can be due to obvious differences in the condensation behavior and in the leaching behavior of minerals for Ca and Cr. Our new data agree with some of the results by [11] that primitive meteorites contain small anomalies in ^{48}Ca . In conclusion, the anomalies in ^{54}Cr and ^{48}Ca in the Murchison samples support the idea that the high density SNIa represent the stellar source for the precursors of these materials. The possible effects in ^{46}Ca would be consistent with s-process ejecta from massive stars [12]. Further work is needed to establish the nature of the carrier of ^{48}Ca anomalies.

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