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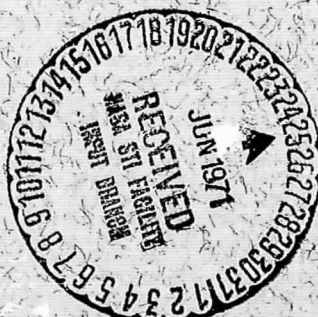
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# THE PRODUCTION RATE OF $\text{Al}^{26}$ FROM TARGET ELEMENTS IN THE BRUDERHEIM CHONDRITE

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**JUNE 1971**



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THE BRUDERHEIM CHONDRITE

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## ABSTRACT

An 840-gram specimen of the Bruderheim chondrite was subjected to magnetic and heavy liquid mineral separation procedures, resulting in a number of chemically distinct samples. These samples were analyzed for cosmogenic  $\text{Al}^{26}$  by non-destructive gamma-gamma coincidence counting. The observed  $\text{Al}^{26}$  specific activities were correlated with the chemical composition of potential target elements by a weighted least squares fitting technique. The calculated  $\text{Al}^{26}$  production rates, in dpm per gram of target element, are: Al,  $1.09 \pm 0.17$ ; Si,  $0.270 \pm 0.009$ ; S,  $0.142 \pm 0.015$ . Production rates from Mg, Ca and NiFe were assumed to be 0, 0, and 0.0022 dpm per gram, respectively.

Most meteorite classes show a flat distribution of  $(\text{Al}^{26})_{\text{obs}} / (\text{Al}^{26})_{\text{calc}}$ , primarily between 0.80 and 1.05 (excluding short exposure age effects). The only exception is the eucrites. The five eucrites with the highest relative  $\text{Al}^{26}$  activities have only  $0.76 \pm 0.04$  of their respective calculated activities. Two Apollo 12 samples, from mean depths of 15-20 cm, have approximately 0.78 of the  $\text{Al}^{26}$  activities calculated for their chemical compositions. A depletion in  $\text{Al}^{26}$  in lunar samples shielded from solar radiation is in accord with a reduced cosmic-ray flux near the earth's orbit, relative to that experienced by most meteorites. The  $\text{Al}^{26}$  depletion in the eucrites suggests that they spent a greater proportion of their orbital periods at or within 1 AU than have most meteorites. The similarity in relative  $\text{Al}^{26}$  contents of the lunar samples and the eucrites may not be a coincidence.



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# THE PRODUCTION RATE OF $\text{Al}^{26}$ FROM TARGET ELEMENTS IN THE BRUDERHEIM CHONDRITE

## INTRODUCTION

In order to use the specific activity of  $\text{Al}^{26}$  in stone meteorites for the estimation of shielding effects, terrestrial ages, short cosmic-ray exposure ages, and effects caused by variations in a meteorite's radiation environment, some knowledge of the effect of chemical composition on the production rate of  $\text{Al}^{26}$  is needed. Because of experimental difficulties in producing and measuring long-lived isotopes in artificial bombardments, the production cross-sections for  $\text{Al}^{26}$  from Al, Si, Mg, S, Ca, and Fe are not known. Unfortunately, while effective cross-sections of long-lived products can be inferred from the production of shorter-lived species in the case of an iron target, the abundance of target elements and the absence of a suitable chain of measurable products prohibits such inferences in the case of stone meteorites.

One approach to the problem (Fuse and Anders, 1969; Heymann and Anders, 1967) involved the measurement of  $\text{Al}^{26}$  in a number of stone meteorites of varying chemical compositions. Low  $\text{Al}^{26}$  activities attributable to short exposure ages or to shielding effects were excluded, and the remaining data were subjected to weighted least squares analyses to determine target element contributions to the production of  $\text{Al}^{26}$ . This method assumes a constant shielding effect, and the same cosmic-ray environment, for all samples studied. Fuse and Anders (1969) estimated production rates of 2.2 dpm  $\text{Al}^{26}$ /kg Fe + Ni, from iron meteorite data, and 7 dpm/kg Ca, from spallation theory, using the preceding Fe + Ni contribution. They then calculated production rates of  $476 \pm 54$  dpm/kg Al,  $310 \pm 10$  dpm/kg Si, and  $38 \pm 44$  dpm/kg S, with the Mg contribution proving incalculable, and estimated to be zero for the other calculations.

In this paper, the contributions of various target elements to the production of  $\text{Al}^{26}$  in a single specimen of the Bruderheim stone meteorite are reported. This approach removes any uncertainty about the shielding and cosmic-ray environment of the samples studied. On the other hand, the calculated relationships are, strictly speaking, only valid for the particular shielding circumstances of this sample, and the cosmic-ray flux to which Bruderheim was exposed. Numerous fragments of the Bruderheim fall have been analyzed radio-chemically (Fireman and DeFelice, 1961; Honda et al., 1961; Rowe and Van Dilla, 1961; and others), with no indication of an unusual cosmic-ray flux. The composite  $\text{Al}^{26}$  activity for the present sample (66 dpm/kg) indicates a well-developed secondary flux, and does not suggest any unusual degree of shielding. Thus the production rates calculated herein should be generally applicable to most stone meteorites.

## SAMPLE PREPARATION

The fusion crust from an 840-gram fragment of Bruderheim was removed (with some effort) with a steel hammer and chisel. The same tools were used to break the sample into approximately one inch pieces, which were further reduced to millimeter-size chips in a steel percussion mortar. The chips were ground with a diamondite mortar and pestle, and the powder shaken in a stack of 100-, 200-, and 400-mesh nylon sieves. The material finer than 400 mesh (about 5% of the total) was put aside. A magnetic portion was removed from each fraction with a hand magnet, and coarse material was repeatedly reground until everything passed at least the 100-mesh sieve.

The MAGNETIC fraction (about 67 grams) was counted for  $\text{Al}^{26}$ , analyzed chiefly by atomic absorption, and further purified in an ultrasonic cleaner. The final product (46 grams, referred to hereafter as the METAL fraction) contained about 6% silicate, versus about 20% in the MAGNETIC fraction. The METAL was also counted for  $\text{Al}^{26}$  and analyzed chemically.

The 100-200 and 200-400 mesh portions (after removal of metal) were shaken separately in methylene iodide (specific gravity =  $3.33 \text{ g/cm}^3$ ) in one liter separatory funnels, and the "sink" fractions were drawn off. X-ray diffraction spectra of the "float" and "sink" fractions indicated a poor feldspar separation in the 100-200 mesh sample. This portion was ground to 200-400 mesh, raked again with a hand magnet, and the heavy liquid separation was repeated. The combined "float" fraction was shaken repeatedly with methylene iodide until no appreciable "sink" fraction was obtained. This sample, 57.5g, referred to as FELDSPAR, was analyzed chemically, and counted for  $\text{Al}^{26}$ .

The heavy material from the methylene iodide separation was passed through a Frantz Isomagnetic Separator at low current to remove strongly magnetic grains. X-ray diffraction examination of this separate indicated primarily metal-olivine aggregates, with some identifiable schreibersite. The remaining material was passed through the Frantz at 0.30 amp,  $30^\circ$  forward slope,  $20^\circ$  side slope, and the magnetic portion was raked repeatedly at this setting until no significant non-magnetic material appeared. X-ray diffraction indicated that the magnetic fraction was enriched in olivine; this 95-gram fraction is henceforth called OLIVINE-1. Raising the current to 0.38 amp produced no additional magnetic material from the non-magnetic fraction.

A second olivine-rich fraction, OLIVINE-2, 191g, was removed at 0.60 amp,  $30^\circ$  forward slope,  $25^\circ$  side slope, at a moderate vibration rate. The portions were raked in counter-current fashion until no appreciable separations were obtained. The 117-gram non-magnetic fraction was enriched in pyroxene, with

some feldspar, and was labeled PYROXENE-1. After counting and chemical analysis, further mineral separations were carried out on the olivine and pyroxene fractions.

The three mixtures were treated separately and repeatedly with Clerici solution having a specific gravity of 4.25. The combined heavy fraction consisted of 26.5 grams rich in TROILITE.

The three troilite-free samples were allowed to settle in distilled water; the fine material which had not yet settled after two minutes was decanted and put aside. The remaining material was separated, using Clerici solutions with specific gravities between three and four, into OLIVINE-3 (101 grams), PYROXENE-2 (47 grams), and PYROXENE-3 (31 grams). Further details concerning the techniques used in the heavy liquid separations are given by Schoen and Lee (1964).

## COUNTING TECHNIQUES

The separated samples were measured non-destructively in a low-level gamma-gamma coincidence counting system, details of which can be found elsewhere (Cressy, 1970). Samples and standards were counted in lucite containers placed reproducibly between the two 10.2 x 10.2-cm NaI(Tl) detectors. Calibrations were made using Na<sup>22</sup> and Al<sup>26</sup> standards mixed with 100-mesh dunite powder. Pure dunite powder was used for background measurements. The N. B. S. Na<sup>22</sup> standard has a reported uncertainty of  $\pm 2\%$ . The Al<sup>26</sup> standard, recalibrated in this laboratory relative to the N. B. S. Na<sup>22</sup> source and to a Batelle Northwest Laboratories Al<sup>26</sup> standard, has an uncertainty of  $\pm 5\%$ . The Al<sup>26</sup> activity in each sample was estimated from the 0.51 MeV coincidence with the 0.51 + 1.81 MeV sum peak. As a check, Na<sup>22</sup> and Al<sup>26</sup> were determined by weighted least squares analysis of the coincidence peaks to which both nuclides contribute. (The various Bruderheim separates were counted approximately 10 years after the meteorite's fall. Sodium-22 had decayed by about a factor of 16, enough that it could not be accurately measured nondestructively, although it still constituted about 10% of the prominent 0.51 x 0.51 MeV coincidence peak.) In all cases the weighted least squares Al<sup>26</sup> values agreed with those from the 0.51 x 2.32 MeV coincidence; the latter values were used in the production rate calculations because of the greater confidence in the Na<sup>22</sup>-free data.

## RESULTS

Table 1 lists the elemental compositions of the various separates (analyses by David Nava of this laboratory), followed by the Al<sup>26</sup> specific activities in disintegrations per minute per kilogram of sample (dpm/kg). The second value for

MASS, g	FELDSPAR	OLIVINE-1	OLIVINE-2	OLIVINE-3	PYROXENE-1	PYROXENE-2	PYROXENE-3	MAGNETIC	METAL	TROILITE
COMPOSITION, %										
Fe	52.8	94.8	191	101	117	47.1	31.2	67.3	45.9	26.5
Ni	8.71	16.4	16.6	17.4	18.4	11.0	8.7	68.4	78.9	57.9
Co	0.42	0.68	0.035	0.01	0.015	0.01	0.01	10.3	13.8	0.06
Cu			0.0056	0.0016	0.0059	0.0014	0.0013	0.41	0.58	0.0033
Cr		0.003	0.002		0.005			0.046	0.067	
S	0.41	1.70	0.03	0.133	0.06	0.165	0.226		0.039	2.69
Si	24.63	20.29	19.49	18.51	20.33	24.77	24.17	0.62	0.39	32.5
Mg	13.09	18.09	19.24	22.62	14.11	17.25	14.11	5.10	1.35	0.88
Al	3.74	0.84	0.65	0.106	1.25	0.37	2.03	3.24	0.83	0.87
Ca	2.55	0.89	1.34	0.22	2.24	1.19	3.85	0.37	0.095	0.085
Na	2.15	0.41	0.36	0.082	0.64	0.23	1.20	0.36	0.086	0.086
K(AA)	0.35	0.054	0.046	0.007	0.093	0.025	0.16	0.22	0.065	0.045
( $\gamma$ )		0.052	0.049	0.009	0.092	0.025	0.14	0.03	0.01	0.01
Mn	0.21	0.32	0.32	0.36	0.26	0.36	0.27	0.02	0.015	<0.02
Ti	0.11	0.10	0.10	0.053	0.11	0.13	0.13	0.06	0.024	0.028
P	0.27	0.08	0.06	0.07	0.17	0.06	0.41	0.17	<0.01	0.12
H <sub>2</sub> O <sup>-</sup>	0.18	0.05	0.06	0.03	0.04	0.12	0.10	0.15	0.034	0.024
TOTAL (0 calc.)	100.8	99.2	100.3	101.1	100.9	99.7	99.4	0.08	0.03	0.02
Al <sup>26</sup> , dpm/kg	104.8 ±7.4	68.3 ±3.2	63.5 ±2.2	50.0 ±4.0	75.5 ±3.3	69.2 ±4.2	91.3 ±6.0	101.2	99.0	98.7
								16.5 ±2.0	4.0 ±3.8	50.0 ±5.3



%K in each column was obtained from measurement of  $K^{40}$  in the non-coincident gamma-ray spectra. The figures for total oxygen were calculated by summing the oxides for all elements; iron as oxide was estimated by correcting the total iron in each separate for the Fe/Ni ratio (5.58) in the metal. The stated uncertainties in the  $Al^{26}$  activities represent one standard deviation, from sample and background counting statistics.

The production rates of  $Al^{26}$  from the principal target elements, Mg, Al, Si, S, Ca, and NiFe, were calculated by weighted least squares analysis. The results of the various computer runs are given in Table 2; negative calculated production rates were forced to zero (see P(NiFe) in row 1). The number of elements taken as unknown is given in the second column. Column three lists the number of measured samples used. The  $Al^{26}$  production rates from the six elements are given next, and the last column lists the calculated chi square value for goodness of fit for each run. The quoted production rate uncertainties reflect both the chi square value and the statistical uncertainties in the measured  $Al^{26}$  activities.

In the first run, all six elements and the  $Al^{26}$  activities in all ten samples were used. For later runs, P(NiFe) and P(S) were estimated independently, and the measured  $Al^{26}$  values were corrected for their contributions. P(NiFe) was estimated from iron meteorites, after Fuse and Anders (1969), at 0.0022 dpm/g NiFe. Production rate from sulfur was taken from run #1. For the remaining runs, the Magnetic, Metal and Troilite samples were omitted, the Magnetic sample because the residual activity after subtraction of P(S) and P(NiFe) was too small to give meaningful production rate information. Thus, the second run used four elements (Si, Al, Mg and Ca) and seven samples.

The interference between silicon and magnesium was always greater than 90%, whereas the interferences between Ca-Si, Ca-Al and Ca-Mg were 15-30%, and the Si-Al and Mg-Al interferences were on the order of 10%. The low calculated magnesium production rates, and the high Si-Mg chemical correlation in these samples, suggest that P(Mg) can be set equal to zero without adversely affecting the results (a feature also noted by Fuse and Anders); this was in fact done in run 3. In run #4, P(Ca) was set equal to zero instead in view of the high calculated uncertainties for P(Ca) and the appreciable Ca correlations. In run #5 both P(Ca) and P(Mg) were set to zero. The chi square value for this last run was lower than those for the earlier runs, indicating that, for these samples at least, the best fit between calculated and observed  $Al^{26}$  activities is obtained when Ca and Mg are assumed to have no contribution to  $Al^{26}$  production. The production rates in this last row are used throughout the remainder of this paper.

Table 2

Al-26 Production Rates in Bruderheim, dpm/g Target Element

Run	Elements	Samples	P(Al)	P(Si)	P(Ca)	P(Mg)	P(NiFe)	P(S)	Chi Square
1	6	10	$1.020 \pm 0.290$	$0.226 \pm 0.052$	$0.218 \pm 0.259$	$0.038 \pm 0.046$	0	$0.142 \pm 0.015$	0.440
2	4	7	$1.060 \pm 0.288$	$0.237 \pm 0.053$	$0.158 \pm 0.261$	$0.026 \pm 0.046$	0.0022	0.142	0.425
3	3	7	$1.010 \pm 0.262$	$0.267 \pm 0.012$	$0.094 \pm 0.226$	0	0.0022	0.142	0.389
4	3	7	$1.147 \pm 0.242$	$0.255 \pm 0.042$	0	$0.015 \pm 0.040$	0.0022	0.142	0.397
5	2	7	$1.090 \pm 0.169$	$0.270 \pm 0.009$	0	0	0.0022	$0.142 \pm 0.015$	0.339

Table 3 compares the observed  $\text{Al}^{26}$  activities in each of the separated Bruderheim fractions with the calculated activities using the production rates from run 5 in Table 2, and from Fuse and Anders (1969). The largest discrepancy in calculated  $\text{Al}^{26}$  activities is seen for the troilite-rich sample; Fuse and Anders had no samples as rich in sulfur for their study. In general, the Fuse and Anders equations yield a high  $\text{Al}^{26}$  activity when the Si/Al ratio is high, and a low activity when the Si/Al ratio is low.

Table 3  
Calculated and Actual  $\text{Al}^{26}$  Activities in  
Bruderheim Fractions, dpm/kg sample

SAMPLE	OBSERVED	A	B
Feldspar	$104.8 \pm 7.4$	94.8	108.1
Olivine-1	$68.3 \pm 3.2$	68.0	66.7
Olivine-2	$63.5 \pm 2.2$	64.6	62.4
Olivine-3	$50.0 \pm 4.0$	59.4	51.9
Pyroxene-1	$75.5 \pm 3.3$	71.7	77.1
Pyroxene-2	$69.2 \pm 4.2$	79.0	71.4
Pyroxene-3	$91.3 \pm 6.0$	85.2	88.0
Magnetic	$16.5 \pm 2.0$	19.3	20.4
Metal	$4.0 \pm 3.8$	6.2	7.3
Troilite	$50.0 \pm 5.3$	16.8	50.7

A. Fuse and Anders, 1969;  $S(\text{Al}^{26}) = 4.8 [\text{Al}] + 3.1 [\text{Si}] + 0.38 [\text{S}] + 0.07 [\text{Ca}] + 0.022 [\text{Ni+Fe}]$

B. Row 5, Table 2:  $S(\text{Al}^{26}) = 10.9 [\text{Al}] + 2.70 [\text{Si}] + 1.42 [\text{S}] + 0.022 [\text{Ni+Fe}]$

## DISCUSSION

Table 4 lists reported  $\text{Al}^{26}$  specific activities in a number of chondrites and lunar samples, together with  $\text{Al}^{26}$  values calculated from Fuse and Anders (1969) and from the elemental production rates chosen in Table 2. Where two or more different  $\text{Al}^{26}$  activities have been reported for a given meteorite, the highest value is given in Table 4, under the presumption that the calculated production rates refer to maximum levels of  $\text{Al}^{26}$ . No effort was made to produce a complete listing of all chondrites for which  $\text{Al}^{26}$  data are available; the selection was intended to be representative of samples having no serious shielding factor or short cosmic-ray exposure age.

It is apparent that, although the production rates reported herein yield generally slightly higher expected  $\text{Al}^{26}$  activities than those of Fuse and Anders, there is little difference between the activity calculations for most chondrites. The largest differences occur for carbonaceous chondrites, which often have lower Si/Al ratios than ordinary chondrites. The calculated activities attributed to the Fuse and Anders production rates may differ somewhat from those reported in Fuse and Anders (1969); the chemical analyses used by these authors were not given, and, for the sake of consistency, their production rates were applied to the chemical analyses taken for this paper.

In the case of McKinney, two different chemical analyses were used for  $\text{Al}^{26}$  calculations. An old analysis reported by Wiik (1969), included an unusually high aluminum concentration of 1.80%, leading to the higher calculated activities (66.9 and 72.8). A more recent analysis reported by Von Michaelis et al., (1969) differed primarily in an aluminum content of 1.12%, yielding the lower  $\text{Al}^{26}$  estimates in each column.

The Apollo 12 samples listed in Table 4 were shielded by at least 8 cm of lunar material. At this depth their  $\text{Al}^{26}$  activities should reflect cosmic-ray production only, in  $2\pi$  geometry; thus the production rates calculated for these samples were divided by two. Samples 12002, 97 and 12053, 42 are bottom surfaces of these rocks, some 10.5 and 8.2 cm respectively from the top surfaces. The agreement between observed  $\text{Al}^{26}$  activities and those calculated from this paper is very good, but must be regarded suspiciously. The extent to which these bottom surfaces may have been exposed to solar radiation while lying on the surface, rather than buried in the soil, is not known. Rock 12034, recovered from a trench at 10-20 cm depth, and the 13-27 cm portions of the double core sample 12028, are better cases for comparison.

The observed  $\text{Al}^{26}$  activity in 12034 is 82% of that predicted from the production rates reported herein. No published chemical analyses are available for the core sample; the expected activity in Table 4 was calculated using the average composition of Apollo 12 fines. The measured  $\text{Al}^{26}$  level in 12028 is

Table 4  
Al<sup>26</sup> in Chondrites and Lunar Samples

SAMPLE	CLASS	Al <sup>26</sup> , dpm/kg		
		OBSERVED	A	B
Lost City	H5	62 ± 1 a	59.8	61.7
Bath	H4	34 ± 4 b	62.9	67.6
Plainview	H5	55 ± 2 b	59.0	60.8
Dwaleni	H5	57 ± 3 a	60.3	62.8
Pribram	H5	59 ± 3 a	60.9	63.0
Bruderheim	L6	60 ± 6 c	64.3	65.7
McKinney	L4	71 ± 5 b	66.9, 63.6	72.8, 65.4
Tathlith	L6	59 ± 3 a	64.5	67.0
St. Séverin	LL	61 ± 5 d	66.3	68.4
Hamlet	LL4	64 ± 6 b	67.6	70.5
Jelica	LL6	68 ± 4 b	64.0	65.3
Manbhoom	LL6	70 ± 5 d	67.4	71.3
Abee	E4	63 ± 7 b	59.3	63.3
Daniel's Kuil	E6	60 ± 5 b	62.2	63.7
Indarch	E3	61 ± 3 d	58.0	61.4
Allende	C3	61 ± 2 e	58.9	64.1
Ornans	C3	53 ± 4 b	56.2	60.6
Felix	C3	54 ± 4 b	58.7	63.5
Alais	C1	44 ± 6 b	38.9	48.2
Orgueil	C1	41 ± 3 b	38.1	44.4
#12002, 97 10.5 cm		52 ± 8 f	42.2	50.0
#12053, 42 8.2 cm		58 ± 8 f	47.2	58.5
#12034 10-20 cm, rock		60 ± 3 f	53.8	73.4
#12028 13-27 cm, core		48 ± 3 f	~47	~65

A. Fuse and Anders, 1969.

B. Row 5, Table 2

a. this laboratory; Tathlith published earlier, corrected here for Al<sup>26</sup> recalibration.

b. Heymann and Anders, 1967.

c. Honda et al., 1961.

d. Fuse and Anders, 1969.

e. Rancitelli et al., 1969.

f. Rancitelli et al., 1971.



approximately 74% of that predicted from the Bruderheim production rates. This ~20% discrepancy suggests that Bruderheim (and most chondrites) may have seen a proportionally larger average cosmic-ray flux than that incident on the moon, in qualitative agreement with the positive cosmic-ray gradient (or, at least, increase) evidenced by the  $\text{Ar}^{37}/\text{Ar}^{39}$  ratios in the metal phase of the Lost City meteorite (Fireman and Spannagel, 1970).

The "gradient" inferred from the  $\text{Ar}^{37}/\text{Ar}^{39}$  measurements is defined by only two points: near 1 AU, where most of the  $\text{Ar}^{37}$  was produced, and 1.8 AU, the time-average distance of Lost City's orbit from the sun, which presumably accounts for the time-average production of 269-year  $\text{Ar}^{39}$ . The inferred increase in the cosmic-ray flux, at average solar modulation, from 1.0 to 1.8 AU is ~60%, leading to a gradient of ~80% per AU. If this gradient is real, then several conclusions can be drawn.

1. The observed  $\text{Al}^{26}$  activity in Lost City, like the  $\text{Ar}^{39}$ , reflects an average production rate at 1.8 AU, but for a much longer time.
2. Lunar samples which have been shielded from solar radiation should have about 60% of the  $\text{Al}^{26}$  activity that would be produced at 1.8 AU. The two samples mentioned above have in fact an average of 78% of the calculated levels of  $\text{Al}^{26}$ .
3. Meteorites having greater average distances from the sun than 1.8 AU should have more  $\text{Al}^{26}$  activity than Lost City, while those with average distances less than 1.8 AU from the sun should have less  $\text{Al}^{26}$  than Lost City.

Pribram, like Lost City, was also photographed during passage through the atmosphere, and its orbit was determined. It spent only 20% of its orbital period inside 1.8 AU, and its average distance from the sun was 3.1 AU. Calculating the fraction of its orbital period which Pribram spent at various radial distances from the sun, and assuming a positive, linear, cosmic-ray gradient of 80% per AU, Pribram should have some 50% more  $\text{Al}^{26}$  activity than Lost City. A cosmic-ray gradient of +30% per AU can be calculated (for average solar modulation) from a comparison of  $\text{Al}^{26}$  contents in Lost City and lunar samples. For a gradient of this size, Pribram should have 30% more  $\text{Al}^{26}$  than Lost City. In fact, the ratio of  $(\text{Al}^{26})_{\text{obs}}/(\text{Al}^{26})_{\text{calc}}$  for Pribram to that for Lost City is  $0.93 \pm 0.07$ , very near unity and, ignoring the possibility that all the Pribram fragments were heavily shielded, does not support any gradient of more than about  $\pm 10\%$  per AU for the energy range of importance to  $\text{Al}^{26}$  production. This energy is certainly not greater than the 1 GeV estimate for  $\text{Ar}^{37}$  and  $\text{Ar}^{39}$  production from Ni and Fe, and at lower energies the effect of solar modulation should increase, yielding a larger gradient.

The  $\text{Ar}^{37} - \text{Ar}^{39}$  data, and the  $\text{Al}^{26}$  differences between the moon and meteorites, argue for some change in the cosmic-ray flux. Let us assume an equally unlikely model - that the cosmic-ray flux changes rather abruptly somewhere between 1 and, say, 1.8 AU. The "point" at which the flux changes is calculable from the known orbits of Pribram and Lost City, with the constraint that each must have spent the same fraction of its orbital period inside this distance, in order to have essentially the same levels of  $\text{Al}^{26}$ . The calculated distance is 1.02 AU, within which each meteorite spent 8% of its orbital period. Since the orbital elements of Lost City and Pribram have certainly changed during the last two million years, the relative amounts of shielding of Lost City and Pribram are unknown, and the  $\text{Al}^{26}$  measurements themselves have standard deviations of several per cent, the value of 1.02 AU must have some uncertainty associated with it in addition to the limiting case assumption of sudden change in flux. But a general conclusion can be drawn. A relatively abrupt increase in cosmic-ray flux beyond  $\sim 1.0$ - $1.1$  AU from the sun can account for the  $\text{Ar}^{37}/\text{Ar}^{39}$  ratios in Lost City and Pribram, and the reduced  $\text{Al}^{26}$  levels in moderately shielded lunar samples. This "abrupt" increase could be a non-linear effect of the interaction of solar radiation with galactic cosmic rays of the energies of interest, resulting in no modulation at all at these energies somewhere outside of  $\sim 1.1$  AU from the sun. The implication that most chondrites spend less than about 10% of their orbital periods inside 1.0-1.1 AU is not startling.

Table 5 compares calculated and observed  $\text{Al}^{26}$  activities in achondrites having long exposure ages. It is apparent that the production rates reported herein differ seriously from those of Fuse and Anders (1969) for many of these meteorites. The latter production rates agree well with observed activities, not surprisingly, since these achondrites were used in the derivation of the Fuse and Anders production rates. The primary cause of the differences between the two sets of calculations is the widely varying Al/Si ratio among these meteorites. Some of the chemical analyses are very old; new analyses might very well change some of the predicated  $\text{Al}^{26}$  levels.

The ratio of observed to calculated  $\text{Al}^{26}$  for Nakhla is 1.13. If Lafayette's composition is anything like that of Nakhla, its  $\text{Al}^{26}$  activity ( $87 \pm 3$ , Fuse and Anders, 1969) will yield a similarly high ratio. Since Nakhla has a 10:1 Ca:Al elemental ratio, it is possible that, in the absence of a calculated production rate from calcium, the expected  $\text{Al}^{26}$  activity is being underestimated. Alternatively, it is possible that Nakhla (and presumably Lafayette) have been exposed to a higher cosmic-ray flux than that experienced by most meteorites, although the mechanism for such an exposure is hard to envision in view of the earlier discussion of  $\text{Al}^{26}$  in Lost City and Pribram.

The calculated  $\text{Al}^{26}$  activity in Norton County is also less than the observed activity; in this case the discrepancy could well be due to uncertainties in the chemical analyses used for the production rate calculations.

Table 5

Al<sup>26</sup> in Achondrites

SAMPLE	CLASS	Al <sup>26</sup> , dpm/kg		
		OBSERVED	A	B
Bishopville Pena Blanca Spring Norton County	AU	83 ± 3 d 65 ± 4 d 79 ± 4 h	87.2 84.5 79.1	82.0 75.5 71.3
Nakhla	N	82 ± 4 d	76.5	72.3
Shalka Johnstown Roda	D	66 ± 3 d 67 ± 2 d 59 ± 3 d	78.0 81.7 80.7	70.5 76.7 81.1
Frankfort Pavlovka	HO	84 ± 4 d 63 ± 2 d	85.1 90.4	92.4 100.9
Angra dos Reis	AN	97 ± 5 d	87.3	106.5
Novo Urei Goalpara	U	46 ± 2 d 39 ± 4 d	59.5 62.1	54.2 57.9
Juvinas Pasamonte Sioux County Padvarninkai Stannern Petersburg	EU	106 ± 4 d 106 ± 4 d 100 ± 5 d 96 ± 4 d 96 ± 12 g 61 ± 3 d	105.4 102.6 105.2 96.3 103.9 100.9	137.0 132.6 137.1 121.2 133.1 128.1

A. Fuse and Anders, 1969.

B. Row 5, Table 2.

d. Fuse and Anders, 1969.

g. Rowe et al., 1963.

h. Herzog and Anders, 1971.

The distributions of  $(Al^{26})_{obs} / (Al^{26})_{calc}$  for the achondrites and other meteorite classes are given in Figure 1. One should expect most meteorites in each class to cluster around a ratio of  $1.0 \pm \sim 0.1$ , with some tailing toward smaller ratios because of shielding effects. The frequency distributions in Figure 1 support this expectation for chondrites and the bulk of the achondrites. But the eucrites appear to be systematically depleted in  $Al^{26}$ .

Juvinas, Pasamonte, Sioux County, Padvarninkai and Stannern have an average  $(Al^{26})_{obs} / (Al^{26})_{calc}$  of 0.763, with an unweighted standard deviation of  $\pm 0.036$ . Stannern (ratio of 0.721) may be somewhat low because of a systematic error (noted in Heymann and Anders, 1967) in the  $Al^{26}$  measurements reported by Rowe et al., (1963).

If this depletion in  $Al^{26}$  is real, it certainly can not be explained by short cosmic-ray exposure age; only Pasamonte has an age as low as five million years, sufficiently long for  $Al^{26}$  to reach equilibrium. A two-stage irradiation affecting all these meteorites equally seems highly unlikely. Likewise, it is difficult to accept the possibility that each sample experienced identical, and considerable, cosmic-ray shielding in space. However, another possible explanation exists. It has been an underlying assumption of studies of long-lived and stable cosmic-ray products in meteorites that the long-term average cosmic-ray flux experienced by all meteorites has been the same. The agreement of observed and predicated  $Al^{26}$  activities in most stone meteorites indicates that this is a reasonable assumption in most cases. However, the  $Ar^{37} - Ar^{39}$  evidence cited earlier for a spatial variation in the cosmic-ray flux, and the reduced  $Al^{26}$  activities in lunar samples, suggest that a meteorite spending most of its orbit near 1 AU might have a lower  $Al^{26}$  activity than that calculated from the production rate data. If this is the cause of the depleted  $Al^{26}$  activities in the eucrites, one may further wonder whether it is only a coincidence that eucrites and the shielded lunar samples display about the same degree of "unsaturation" in  $Al^{26}$ . It is beyond the scope of this paper to elaborate on the discussions of Duke and Silver (1967) and others concerning the possible lunar origin of eucrites, but the  $Al^{26}$  activities in the eucrites do not conflict with such an origin.

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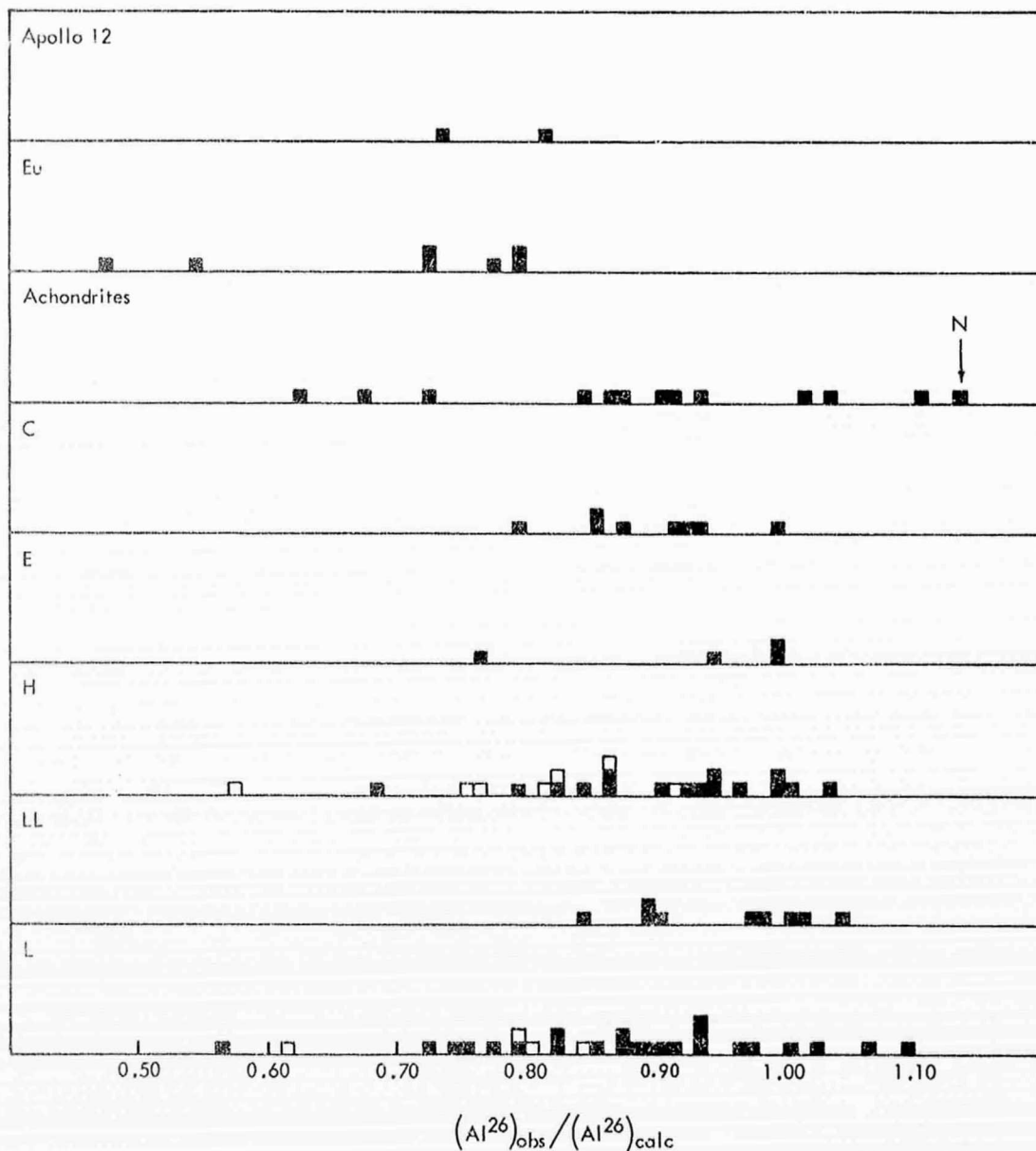


Figure 1.  $(Al^{26})_{obs} / (Al^{26})_{calc}$  in meteorites and two shielded lunar samples. The samples are grouped by class, except that the eucrites have been removed from the general class of achondrites. Meteorites of obvious short cosmic-ray exposure age have been omitted from the figure. The open squares denote  $Al^{26}$  analyses reported by Rowe et al., (1963); these may be systematically low. In the achondrite distribution, "N" refers to Nakhla.



## REFERENCES

- Cressy, P. J. (1970) Multiparameter analysis of gamma radiation from the Barwell, St. Séverin and Tatlith meteorites. *Geochim. Cosmochim. Acta* 34, 771-779.
- Duke, M. B. and Silver, L. T. (1967) Petrology of eucrites, howardites and mesosiderites. *Geochim. Cosmochim. Acta* 31, 1637-1665.
- Fireman, E. L. and DeFelice, J. (1961)  $H^3$ ,  $Ar^{37}$ , and  $Ar^{39}$  in the Bruderheim meteorite. *J. Geophys. Res.* 66, 3547-3551.
- Fireman, E. L. and Spannagel, G. (1970) The radial gradient of cosmic rays from the Lost City meteorite. *S.A.O. Preprints in Meteoritics*.
- Fuse, K. and Anders, E. (1969) Aluminum-26 in meteorities - VI. Anchondrites. *Geochim. Cosmochim. Acta* 33, 653-670.
- Herzog, G. F. and Anders, E. (1971) Radiation age of the Norton County meteorite. *Geochim. Cosmochim. Acta* 35, 239-244.
- Herzog, G. F. and Anders, E. (1970) Absolute scale for radiation ages of stony meteorites. Preprint EFI-70-62.
- Heymann, D. and Anders, E. (1967) Meteorites with short cosmic ray exposure ages, as determined from their  $Al^{26}$  content. *Geochim. Cosmochim. Acta* 31, 1793-1809.
- Honda, M., Umemoto, S. and Arnold, J. R. (1961) Radioactive species produced by cosmic rays in Bruderheim and other stone meteorites. *J. Geophys. Res.* 66, 3541-3546.
- Rancitelli, L. A., Perkins, R. W., Felix, W. D. and Wogman, N. A. (1971) Erosion and mixing of the lunar surface from cosmogenic and primordial radionuclide measurements in Apollo 12 lunar samples. *Proceedings of the Apollo 12 Lunar Science Conference*.
- Rancitelli, L. A., Perkins, R. W., Cooper, J. A., Kaye, J. H. and Wogman, N. A. (1969) Radionuclide composition of the Allende meteorite from non-destructive gamma-ray spectrometric analysis. *Science* 166, 1269-1272.
- Rowe, M. W. and Van Dilla, M. A. (1961) On the radioactivity of the Bruderheim chondrite. *J. Geophys. Res.* 66, 3553-3556.

- Rowe, M. W., Van Dilla, M. A. and Anderson, E. C. (1963) On the radio-activity of stone meteorites. *Geochim. Cosmochim. Acta* 27, 983-1001.
- Schoen, R. and Lee, D. E. (1964) Successful separation of silt-size minerals in heavy liquids. U.S. Geological Survey Prof. Paper 501-B, B154-B157.
- Von Michaelis, H., Ahrens, L. A. and Willis, J. P. (1969) The composition of stony meteorites II. The analytical data and an assessment of their quality. *Earth Planet. Sci. Let.* 5, 387-394.
- Wiik, H. B. (1969) On regular discontinuities in the composition of meteorites, *Commentationes Physico-Mathematicae* 34, 135-145.