The Relative Isotopic Abundance of K^{40} in Terrestrial and Meteoritic Samples

D. S. BURNETT, H. J. LIPPOLT,[1] AND G. J. WASSERBURG

California Institute of Technology, Pasadena

Abstract. Fowler, Greenstein, and Hoyle have proposed that the inner solar system was heavily irradiated during its formation. A consequence of this proposal is that sizable differences in meteoritic and terrestrial K^{41}/K^{40} ratios are possible if the fraction of material which was irradiated was different in the two cases. The isotopic composition of potassium was measured by mass spectrometry for nine stone meteorites, silicate from the Vaca Muerta mesosiderite and the Weekeroo Station iron meteorite, and four terrestrial samples. The measured K^{41}/K^{40} ratios were corrected by normalizing the measured K^{39}/K^{41} ratio to the Nier value of 13.47. This normalization procedure approximately cancels out any variations in the isotopic abundance except those due to nuclear processes. Measurements on enriched standards showed that any variations greater than 1% would certainly have been detected, and variations greater than ½% would probably have been detected with replicate analyses. Within these limits, no variations in the K^{40} abundance between the terrestrial and meteoritic samples could be found which could be ascribed to particle irradiation in the early history of the solar system. Small K^{40} enrichments were observed in Norton County, Weekeroo Station, and Vaca Muerta; however, these appear to have been produced during cosmic-ray irradiation by the $Ca^{40}(n, p)$ reaction. The present results set relatively strong limitations on possible mechanisms for the formation of the earth and the meteorites if the idea of a large-scale irradiation in the early history of the solar system is to be retained. Independent of the model of Fowler et al., limits have been placed on any differential uniform irradiation. The implications of the present work on the K-Ar ages of stone and iron meteorites are discussed. The possibility that iron meteorites are considerably older than the solar system as a whole appears unlikely.

1. INTRODUCTION

The isotopic abundance of K^{40} in meteorites was previously investigated by *Suess* [1939] and *Schumb et al.* [1941] by comparing the specific activity of K in terrestrial and meteoritic samples. These authors were the first to recognize that differences in the isotopic abundance of K^{40} could be used to measure differences in the time of the last event of nucleosynthesis which had contributed new elements to different samples of material. They referred to this as 'differences in the age of the elements.' Their insight was significant because, at that time, many meteorites were thought to come from outside the solar system. Limits of 6% an 3%, respectively, on any variations were set by these authors. *Rik and Shukolyukov* [1954] quote an upper limit of 10% from mass spectrometric determinations. However, before about 1955, K trace analyses were subject to serious contamination errors, and the limits actually set by these experiments are probably at least 10–30%. Meteorites are now regarded as certainly part of our solar system; however, it has recently become apparent that important conclusions of a different sort can be drawn from precise measurements of the relative isotopic abundance of K^{40}.

The purpose of this experiment was to look for variations in the K^{40} abundance due to nuclear reactions that took place during the early history of the solar system. *Fowler et al.* [1962] (FGH) proposed that D and the isotopes of Li, Be, and B were synthesized by the irradiation of meter-sized planetesimals in the region of the terrestrial planets during the formation of the solar system. They assumed for this model that high-energy protons from the sun produced D, Li, Be, and B by spallation reactions in the planetesimals. The planetesimals were assumed to contain H in the form of H_2O; thus neutrons, which were also produced in the spallation reactions, could be effectively thermalized. The thermal neutrons alter the Li and B isotopic ratios produced in spallation by reacting with B^{10} and Li^6 preferentially. The planetesimals

[1] Now at the Max Planck Institut für Kernphysik, Heidelberg, Germany.

were assumed to be large; thus only a fraction q of the planetesimal material would be irradiated ($1/q$ is identical to the dilution factor, F_d, defined by FGH). Using more recent nuclear and abundance data, *Burnett et al.* [1965*a*] calculate the time-integrated neutron flux ψ_n to be 4×10^{21} neutrons/cm^2 and $q = 0.05$ for terrestrial material. There is no a priori reason that the conditions of irradiation would be uniform throughout the early solar system. Consequently, samples of material from different parts of the solar system should show measurable differences in the isotopic composition of certain sensitive elements, even if it is assumed that the irradiated and unirradiated portions of each sample have been thoroughly mixed. At present meteorites are the only samples available for studying isotopic variations between extraterrestrial and terrestrial material.

Isotopic abundances of meteoritic samples have been investigated for several elements from which conclusions can be drawn about the FGH process.

In the FGH calculations the dependence of Li^7/Li^6 on the integrated proton flux is approximately linear for high fluxes [*Burnett et al.,* 1965*a*]. *Krankowsky and Müller* [1964] found the Li^7/Li^6 ratio in 7 meteorites to be within 2% of that found for terrestrial samples. This requires the proton flux for terrestrial and meteoritic material to be the same to within 2% if the FGH model is valid. Thus, in the interpretation of the present experimental results, we shall assume that terrestrial and meteoritic material were exposed to the same flux.

The FGH irradiation would strongly affect the isotopic composition of many elements in the irradiated material. Thus, even if the flux were the same, isotopic variations between terrestrial and meteoritic material could still occur if the relative proportions of irradiated and unirradiated material were not the same, i.e., if there are differences in q. For example, if meteorites were formed from planetesimals that were smaller on the average than those which formed the earth, the fraction of material irradiated would be greater.

Gd, Sm, and Eu have isotopes with very high thermal neutron capture cross sections. For $\psi_n = 4 \times 10^{21}$ neutron/cm^2, these nuclei would be completely destroyed in the irradiated material. Therefore, any variations in isotopic composition only reflect variations in the mixing ratio of irradiated and unirradiated material. *Murthy and Schmitt* [1963] found no variations in Gd, Sm, and Eu in four meteorites, thus it is clear that both terrestrial and meteoritic material cannot be regarded as mostly irradiated or mostly unirradiated material.

Thus, in interpreting the results of the present experiment, we shall assume that both meteoritic and terrestrial material represent a thorough mixture of irradiated and unirradiated material; although the proportions could be variable, the ratio of unirradiated to irradiated material in each case is the vicinity of 20. The analysis of the Murthy-Schmitt experiment by *Burnett et al.* [1965*a*] indicates that the limits set on variations in q are about 20%.

Nuclei, such as K^{40}, which are enhanced strongly in the irradiated material are much more sensitive to variations in q than those which are depleted. For example, the abundance ratio for a depleted nuclide between material which had been irradiated and mixed and material which had never been irradiated is $(1-q)$, which corresponds to a 5% difference. However, the corresponding ratio is infinite for nuclei that were absent in the unirradiated material but synthesized in the irradiated material. For K^{40} the calculations of *Burnett et al.* [1965*a*] indicate that about half of the terrestrial K^{40} would be synthesized in the FGH irradiation, mostly by $K^{39}(n, \gamma)$ but with a small contribution from spallation of iron. Thus the maximum allowed variations in q of 20%, as set by the Gd, Sm, and Eu measurements, affects only one-half of the K^{40} in meteorites. This means that the maximum expected variation in the K^{40} abundance is 10%; or, in general, an $X\%$ variation in q shows up as a $(1/2)X\%$ variation in the K^{40} abundance. Because variations in the K^{40} abundance of 1/2 to 1% can be measured, variations in q can be detected which are a factor of 10 to 20 smaller than would be possible with nuclei that are depleted.

Although there are uncertainties in the estimate of the amount of K^{40} produced, this experiment should remain a sensitive test of variations in q because the calculated maximum variation is an order of magnitude larger than can be measured.

The results of this experiment can be used independently of the FGH model to set limits on the differential primordial irradiation history of the earth and the meteorites. This experiment also has implications for the K-Ar dating of both stone and iron meteorites as well as for other meteoritic isotopic data. A preliminary report of this work is given by *Burnett et al.* [1965*b*].

2. Experimental Procedure

Samples. Three criteria were used in selecting the meteorites investigated. (1) Only meteorites with large amounts of K were chosen in an attempt to avoid K^{40} and K^{41} produced by cosmic-ray irradiation (cosmogenic K^{40} and K^{41}). (2) An attempt was made to sample the principal meteorite classes. (3) When possible meteorites were chosen for which K-Ar ages greater than 4.5×10^9 years had been reported by *Kirsten et al.* [1963].

Samples of Miller, Abee, and Norton County were obtained from Walter Nichiporuk and of Nuevo Laredo from L. T. Silver, both of the California Institute of Technology. Samples of Bruderheim were obtained from R. E. Folinsbee of the University of Alberta; samples of Orgueil, Murray, Pasamonte, Pena Blanca Springs, and Norton County were obtained from the Nininger Meteorite Collection through Carleton Moore of Arizona State University (Tempe); and samples of Weekeroo Station and anorthite ($CaAl_2Si_2O_8$) from Vaca Muerta were obtained from C. Frondel of Harvard University. All of these except Weekeroo Station and Vaca Muerta are falls.

The following terrestrial samples were used as standards: (*a*) a mixture of olivine and hornblende which had approximately the same K concentration (0.1%) as chondritic meteorites, (*b*) plagioclase (about 0.3% K) separated from a granite from the Sandia Mountains, New Mexico, (*c*) Snake River Plateau basalt (0.9% K) obtained from L. T. Silver, (*d*) reagent KCl and KNO_3.

To avoid surface contamination small chips or pieces were used rather than powered samples. These were rinsed briefly in cold 3 *N* HCl. There was no other sample preparation for the stone meteorites.

The silicate inclusions from the Weekeroo Station iron were extracted mechanically. Because surface contamination could be a serious problem in this case, considerable care was taken to obtain clean samples. The specimen was a sawed slab about 1.8 cm thick. Two samples were obtained by two different procedures. The first (Weekeroo I) was picked directly from the sawed surface. The surface was cleaned with acetone, triple distilled water, and 6 *N* HCl. The outer surfaces of the inclusions were removed with a dental burr and the interiors picked out with dental picks and tweezers. The material obtained by this method was highly powdered. The second sample (Weekeroo D) was obtained by sawing thin slices in order to obtain a fresh interior inclusion that did not intersect either surface. This was somewhat difficult because the specimen was thin. The cutting was done with an alundum wheel lubricated with triple-distilled water. The residual alundum on the sawed strips washed off easily, and none could be detected by microscopic examination in the final sample. Weekeroo D was a single interior inclusion except for a small neck (1–2 mm) leading to the surface. The freshly sawed surfaces were cleaned with HCl and water and the inclusion punched out without sampling the material of the neck. A highly magnetic fraction was separated from both samples with a covered magnet, but this did not constitute a thorough magnetic separation. The nonmagnetic portion was used for analysis. The final separate of Weekeroo D was given an additional rinse with 3 *N* HCl. The latter sampling procedure appears to be superior because coarser pieces of silicate material were obtained.

Chemical separation. All samples were dissolved in HF-$HClO_4$ and the K separated from the other major constituents on a 200×15 mm ion-exchange column made from SiO_2 glass using Dowex 50×8 (200–400 mesh) resin. The elution was made with 0.7 *N* HCl. Only platinum, Teflon, and polyethylene apparatus was used in chemical operations in order to avoid the type of contamination by leaching of glassware observed for Sr by *Wasserburg et al.* [1964]. Sample sizes were from 100 to 400 mg. Blanks were measured frequently and ranged from 0.3 to 0.5 μg of K.

Some measurements of K concentrations were made by isotopic dilution using a spike with $K^{41}/K^{40} = 10.0$ and $K^{41}/K^{39} = 13.6$. This was done by spiking an aliquot of the sample after dissolution, allowing both the concentration and isotopic composition of a single sample to be measured. To prevent cross contamination with the highly enriched K^{40} spike, we used a completely distinct set of chemical apparatus for the spiked samples. No evidence of any cross contamination was observed.

Mass spectroscopy. Approximately 10 μg of K was evaporated on a single tantalum filament that had previously been outgassed for 1 hour at a current of 5 amp (bright white heat). A potential of -200 volts was put on focusing plates located directly above the filament in the ion source assembly. The total background K currents from the outgassed filament and the mass spectrometer were less than 10^{-3} of that due to a sample.

The isotopic composition was measured by magnetic scanning on a 12-inch, 60° sector, solid source mass spectrometer. A simple Faraday collector was used to measure the ion currents. The mass spectrometer had never been used for measuring K samples with an anomalous isotopic composition, and it was maintained this way during the course of the experiment. The K isotopic dilution was measured with a different instrument.

The K^{40} peak was well resolved from the tails of the K^{39} and K^{41} peaks (Figure 1). Although the exact shape of the spectrum varied somewhat from measurement to measurement, the K^{40} peak was always well separated. In the early measurements, before November 21, 1964, the base line under the K^{40} peak had a pronounced slope which made interpretation of the zero line subject to some error. This was eliminated by increasing the voltage on the electron repeller plate in front of a collector from -45 to -135 volts, which suggests that the effect was caused by secondary electrons. This modi-

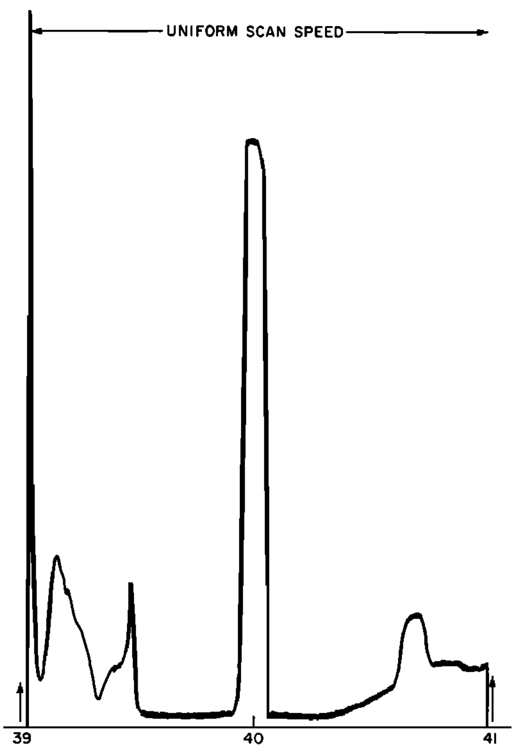

Fig. 1. Uniform scan between masses 39 and 41 showing the K^{40} peak; the base line in the region of the peak and background features are typical of those that were present in all the spectrums. The K^{39} peak is about 10^4 times as large and the K^{41} peak about 580 times as large as the K^{40} peak.

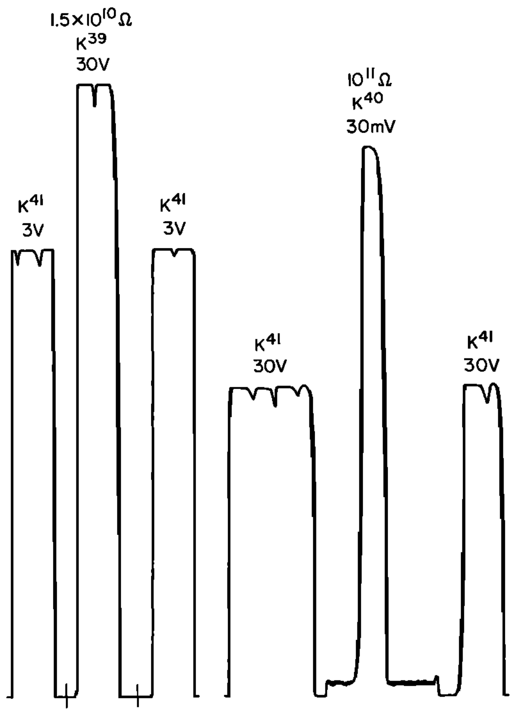

Fig. 2. Illustration of method by which K^{39}/K^{41} and K^{41}/K^{40} ratios were measured. The K^{39}/K^{41} ratio was measured with a 1.5×10^{10}-ohm collecting resistor and the K^{41}/K^{40} ratio with a 10^{11}-ohm resistor. The amplifier output voltage scale on which the data were taken is also shown.

fication gave a significant improvement in the precision of the results. The amplifier scales were calibrated several times during the course of the experiment, and the amplifier and recorder linearity were tested. Possible errors from these sources were found to be negligible.

It was necessary to use two collecting resistors to measure both K^{39} and K^{40} and to obtain a good signal-to-noise ratio for K^{40} (Figure 2). A typical sequence was to measure two K^{41}/K^{40} ratios and then a K^{39}/K^{41} ratio. Results were based on 10–20 K^{41}/K^{40} ratios and 5–10 K^{39}/K^{41} ratios.

Fractionation correction. Because the effect of nuclear processes on the K^{39} and K^{41} abundances in our samples is negligible, the measured K^{39}/K^{41} ratios were used to correct the measured K^{41}/K^{40} ratios for any isotopic fractionation or instrumental discrimination by normalizing the observed K^{39}/K^{41} ratios to the terrestrial value given by *Nier* [1950] of 13.47. Had we used the value of K^{39}/K^{41} = 13.57 given by *Reuterswärd* [1956], all the $(K^{41}/K^{40})_c$ ratios would be shifted to slightly lower values; however, the conclusions of this paper would remain unchanged. The corrected value of the measured K^{41}/K^{40} ratio, which we will denote by $(K^{41}/K^{40})_c$, was then calculated:

$$(K^{39}/K^{41})/13.47 \equiv 1 + \alpha$$

$$(K^{41}/K^{40})_c = (K^{41}/K^{40})(1 + \alpha/2)$$

This correction is necessary because a small but significant amount of mass fractionation usually occurs during the surface ionization process.

Further, this correction procedure essentially removes any variation in the K^{40} abundance due to isotopic fractionation of K in nature by physical or chemical processes; in the present experimental method, therefore, only variations in K^{41}/K^{40} due to nuclear processes would have been detected. No confirmed case of isotope fractionation of terrestrial K by natural processes has been reported. By measuring the specific activity of K^{40} *Mullins and Zerahn* [1948] found no variations in the isotopic abundance to within counting statistics of 0.5%, from which they concluded that any variations in K^{39}/K^{41} were within 1%. Mass spectrometric measurements have set closer limits on K^{39}/K^{41}: *Létolle* [1963] finds no deviation bigger than 0.5%, *Kendall* [1960] reports measurements all within ±0.3%, and *Kaviladze and Abashidze* [1964] within ±0.3%. The question of chemical isotopic fractionation of electropositive elements in meteorites relative to terrestrial matter has been investigated for Ca, which has an 8-mass-unit spread in its isotopes. *Hirt and Epstein* [1964, and to be published] found that any variations were less than 0.2% for 2 mass units.

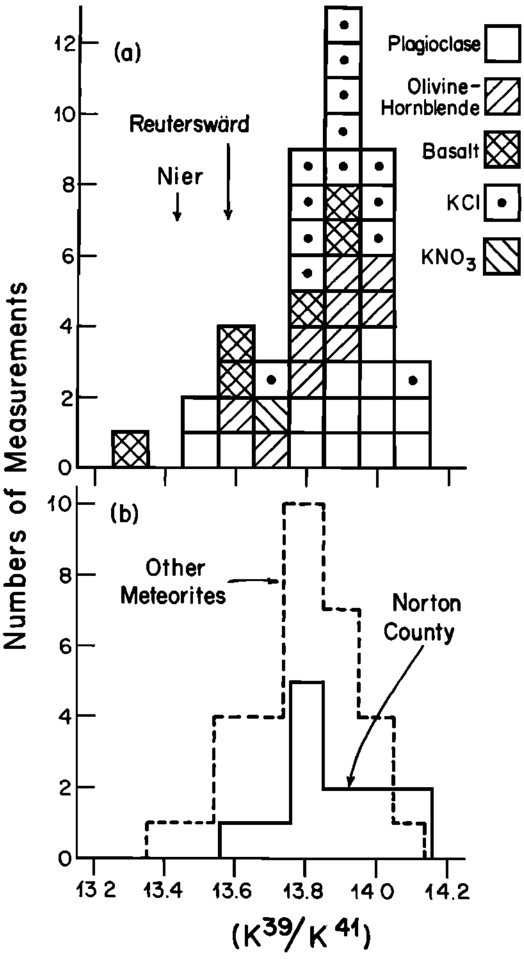

Fig. 3. Distribution of the average K^{39}/K^{41} ratios obtained in various measurements. (*a*) Terrestrial samples. The arrows indicate the absolute values of K^{39}/K^{41} as observed by Nier and Reuterswärd, respectively. (*b*) The solid line is the result of measurements on various samples of Norton County. The dashed line is the results of all other meteorite measurements taken collectively. The total spread of the measurements is about 5%.

K^{39}/K^{41} ratios. Although we made no attempt to investigate possible variations in K^{39}/K^{41}, the measured ratios can be used to give limited support to the assumption that this ratio is constant.

Variations in K^{39}/K^{41} were observed within a single measurement. However, the ratio was usually constant within statistical error under conditions such that the ion current was intense and stable enough to obtain good K^{41}/K^{40} ratios; although occasional variations of up to 1% were observed in a given set of ratios. Only K^{39}/K^{41} ratios taken under the above conditions have been included in the distributions of K^{39}/K^{41} ratios shown in Figure 3. If more than one set of ratios was taken in a single measurement, the K^{39}/K^{41} value from the first set was plotted in Figure 3.

Figure 3*a* shows the distribution in the measured K^{39}/K^{41} ratios for the terrestrial samples. The spread in values for any single sample is as large as the spread of all the samples taken collectively. Thus we conclude that to within statistical error the distribution is the same for all the terrestrial samples, although the basalt ratios tend to be somewhat lower. Figure 3*b* shows the distribution for Norton County, the only single meteorite on which a large number of measurements were made, and the distribution for all other meteorites taken collectively.

TABLE 1. Terrestrial Samples

Sample	Date	$(K^{41}/K^{40})_c$	$(K^{39}/K^{41})_{obs}$	Repeller
Olivine—	Sept. 17, 1964	582 ± 6	13.83	Low
hornblende	Sept. 20, 1964	583.6 ± 3.5	13.90	Low
	Sept. 21, 1964	585.3 ± 2.2	13.90	Low
	Sept. 23, 1964	587.9 ± 1.5	14.00	Low
	Oct. 24, 1964	589.5 ± 3.1	13.76	Low
	Oct. 25, 1964	584.3 ± 3.1	13.98	Low
	Feb. 28, 1965	585.0 ± 1.0	13.88	High
	June 21, 1965	585.1 ± 1.7	13.56	High
Plagioclase	Oct. 30, 1964	587.6 ± 1.9	13.76	Low
(granite	Nov. 5, 1964	585.9 ± 1.5	13.52	Low
Sandia Mts.,	Nov. 7, 1964	584.1 ± 1.6	13.52	Low
New Mexico)	Nov. 19, 1964	585.4 ± 1.5	13.89	Low
	Dec. 3, 1964	586.3 ± 1.8	13.81[a]	High
		583.9 ± 1.0	13.63	High
	Dec. 6, 1964	584.5 ± 1.5	14.04	High
	Dec. 18, 1964	585.0 ± 2.1	13.56	High
	Dec. 20, 1964	585.7 ± 1.9	14.11	High
	May 1, 1965	583.1 ± 1.0	14.01[a]	High
		584.5 ± 1.6	13.88	High
	May 30, 1965	584.3 ± 1.7	13.94	High
	May 31, 1965	582.2 ± 1.8	13.94[b]	High
	June 7, 1965	587.4 ± 1.1	13.97	High
	June 12, 1965	585.6 ± 2.1	14.05[c]	High
	June 12, 1965	584.4 ± 1.5	14.10	High
Snake River	April 11, 1965	585.7 ± 2.0	13.60[c]	High
Plateau basalt		582.9 ± 1.8	13.51	High
		583.7 ± 3.1	13.46	High
	June 2, 1965	584.6 ± 1.7	13.30[c]	High
		584.7 ± 1.8	13.17	High
	June 5, 1965	583.2 ± 1.0	13.69	High
	June 16, 1965	585.5 ± 0.9	13.84	High
	July 17, 1965	586.3 ± 2.0	13.88	High
	July 20, 1965	584.0 ± 1.4	13.55	High
	Aug. 7, 1965	582.5 ± 1.1	13.87	High
KNO_3	Oct. 28, 1964	585.6 ± 2.0	13.65	Low

[a] Made with double-filament ion source.
[b] Rerun of previous sample and filament.
[c] Different sets of ratios taken during same measurements.

Neither of these distributions differs significantly from that of the terrestrial samples.

The data plotted in Figure 3 show that there are no differences in K^{39}/K^{41} among the various samples to within about 2 to 3%. The average of all K^{39}/K^{41} measurements shown in Figure 3 is 13.83. However, the absolute value of the points and the shapes of the distributions in Figure 3 have only relative significance because the measurements were not made under rigidly controlled conditions. Closer limits on variations in K^{39}/K^{41} between terrestrial and meteoritic material could be set by adopting well-standardized procedures, but this was not the purpose of this experiment.

Interfering ions. Although chemical separations were good, we explicitly investigated possible interference by Ca^{40+} or $Mg^{24}O^{16+}$ ions. Samples were analyzed with about equal amounts of K and Ca and of Mg and K with Mg as $Mg(NO_3)_2$. The Ca-K sample gave $(K^{41}/K^{40})_c = 585.4 \pm 1.5$ under normal K running conditions, and the Ca^{40} did not appear until much higher temperatures were used. The Mg-K sample gave 584.5 ± 1.5, and no Mg emission was observed even at temperatures much higher than those at which K was measured. These results agree well with the measurements on pure K samples, which indicates that Ca^{40+} or $Mg^{24}O^{16+}$ interference is negligible.

3. Results and Discussion

Terrestrial samples. The results of the terrestrial K measurements are given in Table 1. Table 1 gives the $(41/40)_c$ ratio (corrected for fractionation), the measured K^{39}/K^{41} ratio, and an indication of whether the measurement was made with a high or low repeller voltage. The quoted statistical errors are the average deviation of a set of ratios. The $(41/40)_c$ ratios are plotted in Figure 4*a*. The value of K^{41}/K^{40} reported by *Nier* [1950] is 581 ± 5. For those samples on which a large number of measurements were made, the distributions shown in Figure 4*a* are identical to well within the spread of the values for a single sample. The only difference between Figures 4*b* and 4*c* is that the low repeller voltage data show a wider spread and tend to have larger statistical errors. The total spread in the uncorrected K^{41}/K^{40} ratios is about 4 to 5% (close to that shown for the K^{39}/K^{41} ratios in Figure 3); the total spread after correction is 1.2% as shown in Figure 4. This shows that the correction procedure used gives consistent results.

The only data which were of apparently good quality that have not been listed in Table 1 and subsequent tables are those for a series of measurements made around April 1, 1965.

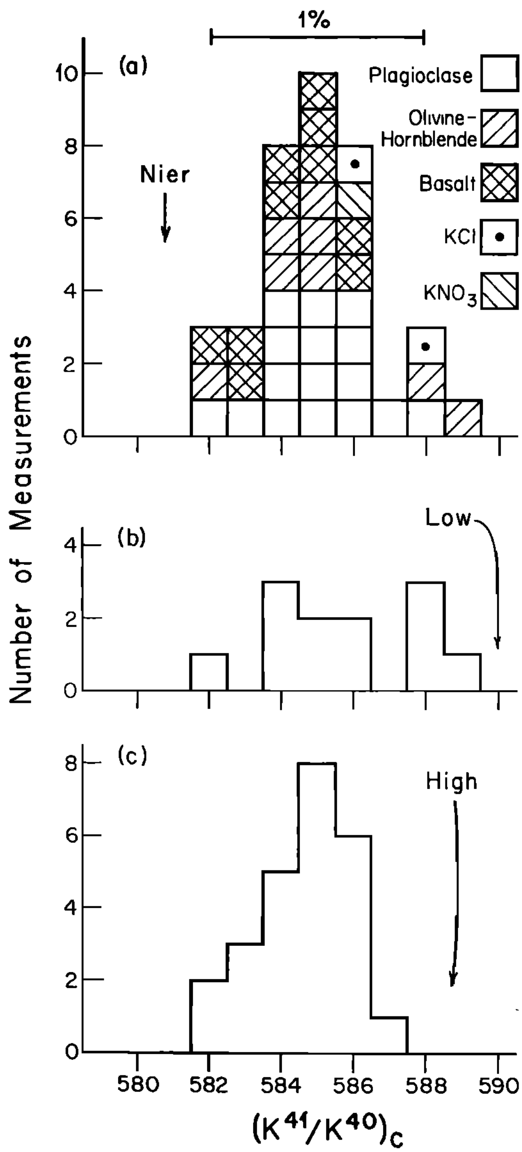

Fig. 4. Distribution of the corrected $(K^{41}/K^{40})_c$ ratios for terrestrial samples: (*a*) distinguishes the results for the various samples; (*b*) and (*c*) show the distributions of the results plotted in (*a*) for measurements made with low and high repeller voltage, respectively.

Some of these measurements showed changes of up to 2% in $(K^{41}/K^{40})_c$ during the course of the measurements; we attributed them to nonlinearity of the 10^{11}-ohm resistor. In subsequent measurements alternate sets of ratios were taken with two different 10^{11}-ohm resistors. As Table 2 shows, the results from the two resistors are in excellent agreement. Moreover, the values for $(K^{41}/K^{40})_c$ agree well with the earlier measurements, which indicates that errors due to similar effects were not significant in the earlier data. Note that these measurements are only sensitive to changes in resistor nonlinearity. A nonlinearity which was constant to within about ½% among the resistors used would have gone undetected. This would have no effect on our relative measurements, but it would affect the absolute values of $(K^{41}/K^{40})_c$.

The measurement of the plagioclase sample on November 19, 1964, was made using several different settings for the collector slit width. The $(K^{41}/K^{40})_c$ ratio was found to be independent of slit width to within the statistical error.

Enriched standards. Sensitivity in detecting abundance variations is usually estimated from statistical errors; however, we have determined it experimentally (a more reliable method) by measuring a series of KCl standards which were slightly enriched in K^{40}. The analyses of a series of standards with enrichments of 0.000, 0.469, 1.295, 1.83 and 3.50% are shown in Table 3. The observed enrichments were calculated by assuming 585 for the terrestrial K^{41}/K^{40} ratio. The 3.5 and 1.8% standards are quantitatively resolved from the 0% standard

TABLE 2. Comparison of $(K^{41}/K^{40})_c$ for Two Different 10^{11}-Ohm Resistors for Some Terrestrial Samples

Sample	Date	Resistor 1	Resistor 3
Plagioclase	May 30, 1965	583.2 ± 1.6	585.3 ± 1.8
	June 12, 1965	584.0 ± 1.7	584.7 ± 1.4
Basalt	June 2, 1965	585.0 ± 1.9	584.3 ± 1.5
	June 5, 1965	583.1 ± 1.0	583.2 ± 1.0
	June 16, 1965	585.3 ± 0.9	585.6 ± 0.8
	July 17, 1965	586.1 ± 1.9	586.5 ± 2.1
	July 20, 1965	584.2 ± 1.4	583.8 ± 1.4
	Aug. 7, 1965	582.3 ± 1.2	582.6 ± 1.0

TABLE 3. Enriched K^{40} Standards

Enrichment, %	Date	$(K^{39}/K^{41})_{obs}$	$(K^{41}/K^{40})_c$	Observed Enrichment, %
0.000	Nov. 21, 1964	13.73	588.1 ± 4.4	−0.5[a]
	Dec. 5, 1964	13.89	586.1 ± 1.5	−0.2
0.469	Feb. 27, 1964	13.88	580.0 ± 1.2	0.9
	May 30, 1965	13.95	581.4 ± 2.0	0.7
	June 1, 1965	13.87	582.0 ± 1.4	0.5
	June 15, 1965	13.81	579.9 ± 1.1	0.9
	June 19, 1965	13.99	582.8 ± 2.0	0.3[b]
	July 18, 1965	13.80	580.9 ± 1.0	0.7
1.295	Nov. 22, 1964	13.82	579.3 ± 2.7	1.0
	May 31, 1965	13.90	576.2 ± 1.4	1.5
	June 22, 1965	13.88	575.0 ± 1.5	1.7
1.83	Nov. 21, 1964	13.79	575.8 ± 2.1	1.5
	Dec. 19, 1964	14.11	574.0 ± 1.9	1.9
3.50	Nov. 22, 1964	13.99	565.3 ± 2.5	3.4

[a] Measured with low repeller voltage; all others measured with high voltage.
[b] Measurement made with double-filament source.

and the other terrestrial samples. The distribution of $(K^{41}/K^{40})_c$ for the terrestrial, 0.5%, and 1.3% standards is plotted in Figure 5. The distribution for the 0.5% standard is distinct but overlaps that of the terrestrial samples; the measurements of the 1.3% standard are clearly resolved. We conclude that any variations of the K^{40} abundance in meteorites of 1% or greater would have certainly been detected, variations of 0.5 to 1% would probably have been detected upon replicate analysis, and variations of less than 0.5% would not have been detected. An uncertainty of 0.5% is about 2 to 3 times the average deviation of ratios taken from a good measurement.

Stone meteorites. The results for some of the stone meteorites are given in Table 4, and the distribution of $(K^{41}/K^{40})_c$ in Figure 6*a* is compared with that of the terrestrial and 0.5% standards shown in Figure 6*b*. The initial measurements of Abee showed a considerable spread, but two later measurements of superior quality gave $(K^{41}/K^{40})_c = 584$. Thus there is no good evidence for any enrichment in Abee. With the exception of two values for Abee, all these stone meteorite data lie within the range of the terrestrial samples, and the K^{40} abundance in these eight stone meteorites is indistinguishable from that of the terrestrial samples to within the limits quoted above.

Norton County. We have excluded data on the Norton County achondrite, Vaca Muerta mesosiderite, and Weekeroo Station iron from the previous discussion. These meteorites show small enrichments in K^{40}. We now wish to discuss these in detail and then give arguments that these enrichments represent recent cosmic-ray production of K^{40} rather than any evidence for nucleosynthesis during the early history of the solar system.

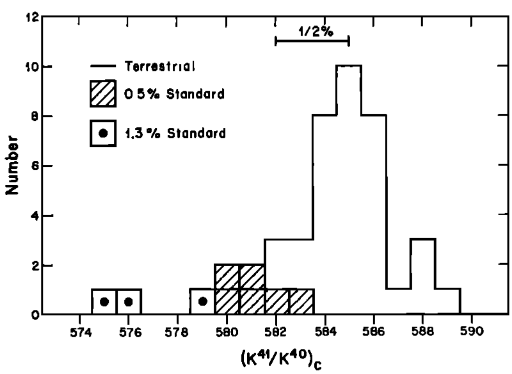

Fig. 5. Comparison of the distributions in $(K^{41}/K^{40})_c$ for the 0.5 and 1.3% enriched standards with that of the terrestrial samples.

The results for Norton County are given in Table 5. The $(K^{41}/K^{40})_c$ ratios are plotted in Figure 7*a* and can be compared with the terrestrial and 0.5% standard distributions plotted in Figure 7*b*. Six samples of Norton County were analyzed, and all but one (number 6) showed enrichments greater than 0.5% that were variable from sample to sample.

To confirm the existence of an enrichment, the identical filament and sample used for the measurement of Norton County 1 on October 29, 1964, was used for a remeasurement with a different mass spectrometer. This instrument was similar to the one normally used except for a poorer vacuum and an electron multiplier detector. An enrichment of 3.2 ± 1.4% was observed.

The spread in $(K^{41}/K^{40})_c$ observed for the Norton County 1 sample is quite large; however, the measurement of September 24, 1964, was much poorer than the later ones, which implies that the lower values are more reliable. All measurements of Norton County 1 were made with the low repeller voltage and are probably less reliable quantitatively than the data on the other Norton County samples.

Samples 1 and 3 were chips obtained from W. Nichiporuk. The remainder came from a single large specimen in the possession of C. Moore. Norton County is largely enstatite ($MgSiO_3$) and contains relatively large single crystals in a matrix of mostly brecciated enstatite [*Beck and La Paz*, 1951; *Keil and Fredriksson*, 1963]. Sample 4 was single-crystal material; samples 5, 6, and 7 were from the brecciated material. The K concentration measurements given in Table 5 were on aliquots of the same samples as were used for the isotopic composition measurements. Small aliquots were taken, which made a blank correction necessary for all samples. The sample weights given in Table 5 are the amounts used for the composition measurements. The K concentrations were found to be quite variable. *Kirsten et al.* [1963] measured 67, 74, and 68 ppm K in three samples of Norton County and *Vinogradov et al.* [1960] reported 230 ppm.

TABLE 4. Meteorites Showing No Enrichments

Meteorite	Classification	Date	$(K^{41}/K^{40})_c$	K^{39}/K^{41}	Repeller	K, ppm
Nuevo Laredo	Eucrite, Ca-rich, pyroxene-plagioclase, or basaltic achondrite	Sept. 25, 1964	587.2 ± 3.1	13.85	Low	367[c]
		Oct. 26, 1964	582.3 ± 2.3	13.91	Low	
		Dec. 4, 1964	582.5 ± 1.8	13.84	High	
Miller (Arkansas)	H-group or olivine-bronzite chondrite	Oct. 1, 1964	587.6 ± 1.7	13.39	Low	720[d]
		Oct. 7, 1964	587.8 ± 1.2	14.00	Low	
		Dec. 19, 1964	585.8 ± 1.0	13.85	High	
Abee	Enstatite chondrite	Oct. 23, 1964	580.8 ± 2.4	13.94	Low	822[d]
		Oct. 28, 2964	579.3 ± 2.0	13.62	Low	
		Nov. 6, 1964	586.5 ± 3.3	13.71	Low	
		Dec. 5, 1964	584.1 ± 1.3	13.80	High	
		June 21, 1965	583.5 ± 1.8	13.97	High	
Bruderheim	L-group or olivine-hypersthene chondrite	Oct. 24, 1964	583.3 ± 2.4	13.76	Low	890[d]
		Oct. 26, 1964	581.9 ± 2.8	13.88	Low	
		Dec. 17, 1964	584.1 ± 1.1	13.63	High	
		June 5, 1965	584.0 ± 1.5	13.75	High	
		June 16, 1965	581.7 ± 1.9	13.72	High[a]	
Orgueil	Type I carbonaceous chondrite	Nov. 5, 1964	587 ± 4	13.86	Low	560[e]
		Nov. 7, 1964	582.4 ± 1.7	13.78	Low	
		Dec. 5, 1964	584.3 ± 1.8	13.56	High	
Murray	Type II carbonaceous chondrite	Nov. 20, 1964	586.1 ± 1.1	13.99	Low	320[e]
		Dec. 7, 1964	583.7 ± 1.8	13.81	High	
Pena Blanca Springs	Ca-poor, aubrite, or enstatite chondrite	Dec. 20, 1964	585.1 ± 1.5	13.76	High	296[d]
		June 4, 1965	585.5 ± 1.6	13.90	High	
Pasamonte	Eucrite, Ca-rich, pyroxene-plagioclase, or basaltic achondrite	April 5, 1965	584.6 ± 1.4	13.55	High	425[c]
		June 3, 1965	582.3 ± 1.4	13.76[b]	High	
			583.2 ± 2.2	13.77		
		June 7, 1965	585.1 ± 1.0	13.83	High	
		June 15, 1965	581.8 ± 0.9	13.69[a]	High	

[a] Rerun of previously measured sample and filament.
[b] Two sets of ratios taken during same measurement.
[c] *Gast* [1965].
[d] *Kirsten et al.* [1963].
[e] *Edwards and Urey* [1955].

The amounts of excess K^{40}, denoted by K^{40*}, shown in Table 5 were calculated from our best estimates of $(K^{41}/K^{40})_c$, which are summarized in Table 6 assuming terrestrial $K^{41}/K^{40} = 585$. For samples 3 and 4, which have low K concentrations, a small blank correction has been applied. A blank of 0.7 μg of K was assumed because these samples were passed twice through the ion-exchange column.

Estimates of cosmogenic K^{40} in Norton County. The small and variable K^{40} enrichments coupled with the high cosmic-ray exposure age of Norton County (2.3×10^8 years by the He^3-H^3 method, *Begemann et al.* [1957] and 5×10^8 years by the Ar^{39}-Ar^{38} method, *Fireman and DeFelice* [1960]) suggest that the K^{40} enrichments may be cosmogenic. We will adopt 2.3×10^8 years for the exposure age in the following discussion.

There are several possible nuclear reactions for the production of K^{40} by cosmic rays. We first consider the spallation of iron-group nuclei which is the source of the cosmogenic K^{40} observed in iron meteorites [*Stauffer and Honda,*

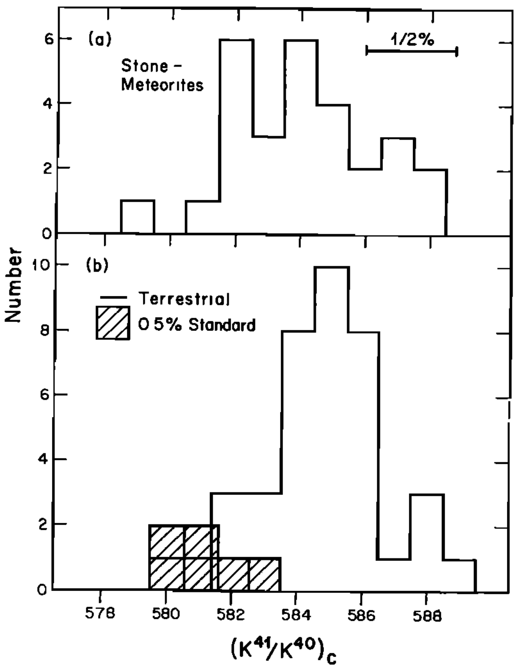

Fig. 6. (*a*) Distribution of $(K^{41}/K^{40})_c$ for those stone meteorites showing no enrichments in K^{40}. (*b*) Distribution for terrestrial and 0.5% standards for comparison.

1962; *Voshage and Hintenberger,* 1963]. We estimate the Fe spallation contribution three ways:

1. The measurements of K^{40} concentrations by Stauffer and Honda and the cosmic-ray exposure ages tabulated by *Anders* [1963] for iron meteorites indicates that roughly 10^{13} atoms K^{40}/g iron per 10^9 yr are produced by cosmic rays. The calculations of *Arnold et al.* [1961] (AHL) indicate that this production rate would not vary much for depths less than 100 g/cm^2. Norton County has about 2% iron-group nuclei [*Beck and LaPaz,* 1951; *Wiik,* 1956]. Thus, this estimate of the amount of K^{40*} produced in Norton County is about 5×10^{10} atoms/g, or a factor of 5 to 30 less than that observed (see Table 5).

2. The only cosmogenic nuclide measured in Norton County that is mainly produced by iron spallation is Ar^{39}, although the $Ca^{40}(n, 2p)$ or $Ca^{40}(\pi^-, \pi^+n)$ reactions could also contribute to the Ar^{39} yield. *Fireman and De Felice* [1960] measured 1 dpm/kg of Ar^{39} in Norton County. AHL calculate the relative production rates of K^{40} and Ar^{39} from Fe spallation to be 1.6, independent of depths less than 100 g/cm^2. This gives $K^{40*} = 1.8 \times 10^{11}$ atoms/g assuming 2.3×10^8 years as the exposure age of Norton County, or a factor of 2 to 7 less than that observed.

3. The direct production rate of K^{40} by Fe spallation calculated by AHL is 35 atoms/min per kg Fe at 100 g/cm^2 depth and 48 atoms/min per kg Fe at 10 g/cm^2. Using the data given above, we get 0.8 to 1.1×10^{11} atoms/g, or a factor of 3 to 20 less than that observed.

Although there is a factor of 4 spread in the above estimates, they all are consistently low, and other reactions must be considered if a cosmogenic origin is to be established. The most likely ones are $K^{39}(n, \gamma)$ and $Ca^{40}(n, p)$. Spallation reactions on the heavier Ca isotopes are not important.

Norton County samples 4–7 have been exposed to about the same cosmic-ray flux because they were all taken from a single large piece. If the excess K^{40*} were due to $K^{39}(n, \gamma)$, the $(K^{41}/K^{40})_c$ ratio should have been constant; however, real variations appear to exist. Further, the required time-integrated neutron flux to produce $K^{40*}/K^{39} \approx 10^{-6}$, as we observe, is 5×10^{17} neutrons/cm^2. This would produce about 6×10^{-8} cm^3 of Ar^{36} (at STP) assuming Cl = 5.5 ppm as quoted by *Begemann and Vilcsek* [1965] on the basis of the work of Van Guten. This is about 3 times the amount of Ar^{36} measured by *Kirsten et al.* [1963]; furthermore, the observed Ar^{38}/Ar^{36} ratio is about 1.5, which is what is expected from spallation. In Norton County Ar^{36} and Ar^{38} were produced primarily by Ca spallation due to the low Fe concentration. Also Begemann and Vilcsek were unable to detect any Cl^{36} activity in Norton County. Thus it appears unlikely that the excess K^{40*} in Norton County is due to $K^{39}(n, \gamma)$.

The large cross section for the Ca^{40} (n, p) reaction (520 mb at 6 Mev, [*Urech et al.,* 1961] suggests that it should be considered as a source of cosmogenic K^{40*}. An estimate of the K^{40*} yield for Norton County has been made by using the shape of the S^{32} (n, p) excitation function [*Allen et al.,* 1957] scaled by a factor 1.7 to match the above Ca^{40} (n, p) cross section

TABLE 5. Meteorites Showing Enrichments

Meteorite and Classification	Sample Weight, mg	Date	K^{39}/K^{41}	$(K^{41}/K^{40})_0$	K, ppm	Ca, %	K^{40*}, 10^{12} atoms/g
Norton County 1	245	Oct. 24, 1964	13.70	575.3 ± 1.1		0.46	
Ca-poor,			13.80	578.4 ± 1.8[a,b]			
enstatite, or		Oct. 29, 1964	13.79	568.9 ± 1.7[b]			
aubrite		Nov. 5, 1964	13.76	571.3 ± 2.2[b,d]			
achondrite[c]							
Norton County 3[c]	433	Nov. 8, 1964	13.57	571.9 ± 3.5[b]			
					23 ± 5	1.27	0.65 ± 0.2
		Dec. 3, 1964	13.81	577.6 ± 1.2			
		Dec. 17, 1964	13.55	577.6 ± 1.7[d]			
		June 22, 1965	14.09	575.4 ± 0.8			
Norton County 4	401	Dec. 19, 1964	14.01	577.3 ± 2.4			
			13.80	578.0 ± 3.0[a]		0.70[f]	
					13 ± 4	0.65	0.37 ± 0.15
		Dec. 22, 1964	13.77	577.1 ± 1.6			
Norton County 5	265	Dec. 18, 1964	13.85	579.7 ± 0.6			
			13.82	579.8 ± 1.9[a]	102 ± 2	2.42	1.6 ± 0.6
		Dec. 20, 1964	13.76	579.6 ± 2.4[d]			
		Dec. 22, 1964	13.97	579.6 ± 1.3			
Norton County 6	121	April 10, 1965	13.88	584.6 ± 2.4			
		April 12, 1965	13.87	584.9 ± 1.3[d]			
					83 ± 3	0.80	0.50 ± 0.50
		June 22, 1965	14.06	580.5 ± 1.4	86 ± 3[e]		
		July 15, 1965	14.09	583.4 ± 1.7[d]			
Norton County 7	207	June 7, 1965	13.66	579.6 ± 1.7[a]			
			13.75	580.9 ± 1.6	61 ± 3	1.25	0.77 ± 0.4
		June 21, 1965	13.75	582.0 ± 1.7			
Weekeroo	274	June 23, 1965	13.72	581.0 ± 2.3	2100	4.8	33 ± 20
Station I (iron;		June 24, 1965	13.84	580.2 ± 1.1			
brecciated		Aug. 9, 1965	13.59	580.0 ± 1.6			
octahedrite)							
Weekeroo	66	July 19, 1965	13.67	580.9 ± 1.1	4400	3.5	
Station D							
			13.73	586.7 ± 0.9[a]			
		July 21, 1965	13.93	585.4 ± 0.6			
		Aug. 8, 1965	14.08	583.7 ± 1.1			
Vaca Muerta	46	July 19, 1965	14.01	576.1 ± 1.4	180 ± 40	13.1	5.8 ± 2
anorthite		Aug. 7, 1965	13.81	575.0 ± 0.8[d]			
(mesosiderite)							

[a] More than one set of ratios taken during measurement.
[b] Measured with low repeller voltage; all others measured with high voltage.
[c] Sample obtained from W. Nichiporuk, all others from C. Moore.
[d] Rerun on previously measured sample and filament.
[e] Two different spikings.
[f] Two different aliquots from same dissolved sample.

at 6 Mev, and a flux spectrum given by

$$\frac{dN}{dE} = \frac{3.7}{E}\left(1 + \frac{0.01}{E} + \frac{1.1 \times 10^{-5}}{E^2}\right) \frac{\text{particles}}{\text{cm}^2 \text{ sec bev}}$$

This is the spectrum used in the AHL calculations at 10-g/cm² depth for the energy region 2–100 Mev. (The energy spectrums for 2–100 Mev in Table 2 of AHL are given incorrectly as ${}^{dN}/dl\ nE$ rather than dN/dE; however, the entries for other energy regions are correct.) The energy range of importance for the Ca^{40} (n, p) reaction is 2–20 Mev with about 80% of the yield occurring below 10 Mev. At these energies the AHL spectrum is composed almost entirely of neutrons. As suggested by the work of *Honda et al.* [1961], we shall assume

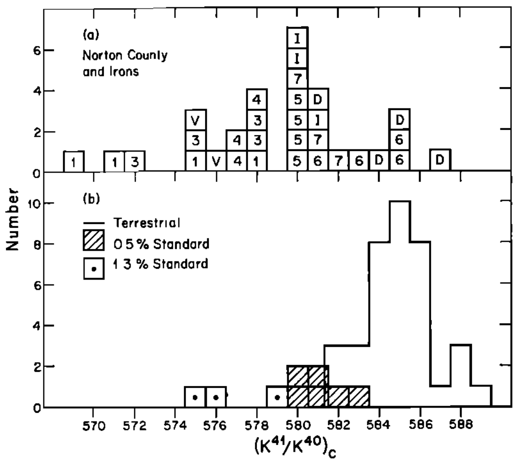

Fig. 7. (*a*) Distribution in $(K^{41}/K^{40})_c$ for meteorites showing an enrichment in K^{40}: Norton County, Weekeroo Station, and Vaca Muerta. The numbers 1–7 refer to the various samples of Norton County listed in Table 5. *I* and *D* refer to the two samples of Weekeroo Station, and *V* refers to Vaca Muerta. (*b*) Distribution of terrestrial, 0.5%, and 1.3% standards for comparison.

that the AHL spectrum derived for irons should also be applicable to stones. This calculation gives $K^{40*} \approx 1 \times 10^{13}$ atoms/g assuming 1% Ca for Norton County, which is about 15 times the average measured K^{40*}. If a depth of 100 g/cm^2 rather than 10 g/cm^2 were assumed, the K^{40*} calculated would increase by about a factor of 1.5. This calculation would imply that far greater enrichments should have been observed. However, as discussed by AHL, the calculated Ni^{58} (n, p) Co^{58} and Fe^{56} $(n, 2n)$ Fe^{55} production rates also appear to be high relative to the observed disintegration rates in the Aroos iron by factors of about 4 and 2, respectively. On the other hand, AHL obtain satisfactory agreement between calculated production rates and the experimental disintegration rates for cosmogenic spallation products produced by reactions of particles above about 100 Mev. Particles of energy less than 10 Mev are more important in the production of K^{40*} from Ca^{40} (n, p) than for the production of either Co^{58} or Fe^{55}. In turn, the Co^{58} production rate is more sensitive to particles in this energy range than that of Fe^{55}. Thus, the above discussion suggests that the AHL spectrum below 100 Mev—taken directly from the measurements of *Hess et al.* [1959] of cosmic ray produced neutrons in the atmosphere—is too steep; and, in particular, the number of particles below 10 Mev is considerably too large. Nevertheless, the above calculation indicates that the Ca^{40} (n, p) reaction should be very effective in producing K^{40*} in a stone meteorite, and is a very likely source for the K^{40*} observed in Norton County.

TABLE 6. Summary of Best Estimates of Measured $(K^{41}/K^{40})_c$ Values

		$(K^{41}/K^{40})_c$[a]
Terrestrial samples	Olivine-hornblende	585
	Plagioclase	585
	Basalt	584
	KCl	586
	KNO_3	586
Meteorites	Nuevo Laredo	583
	Miller	586
	Abee	584
	Bruderheim	584
	Orgueil	583
	Murray	585
	Pena Blanca Springs	585
	Pasamonte	584
	Norton County 1	570
	Norton County 3	576[b]
	Norton County 4	576[b]
	Norton County 5	580
	Norton County 6	583
	Norton County 7	581
	Weekeroo Station I	580
	Weekeroo Station D	584
	Vaca Muerta (anorthite)	575

[a] An uncertainty of ±2 should be assigned to the numbers in this table.
[b] Correction for blank applied.

If the observed K^{40*} enrichments are due to $Ca^{40}(n, p)$, then K^{40*} should be proportional to the Ca concentration because at least four of the Norton County samples have been exposed to about the same cosmic-ray flux. The Ca concentrations listed in Table 5 were measured by isotopic dilution on aliquots of the Ca fraction from the samples that were used for the K^{40} abundance measurements. The Ca^{43} + Ca^{48} spike was provided by S. Epstein. The Ca values are reliable only to about 10% because of uncertainty in chemical yield. Although the errors are large, the plot of K^{40*} versus Ca concentration in Figure 8 suggests that a correlation exists. Although from a different source, sample 3 also appears to have received about the same flux as the other samples. The points on Figure 8 correspond to a K^{40*} production

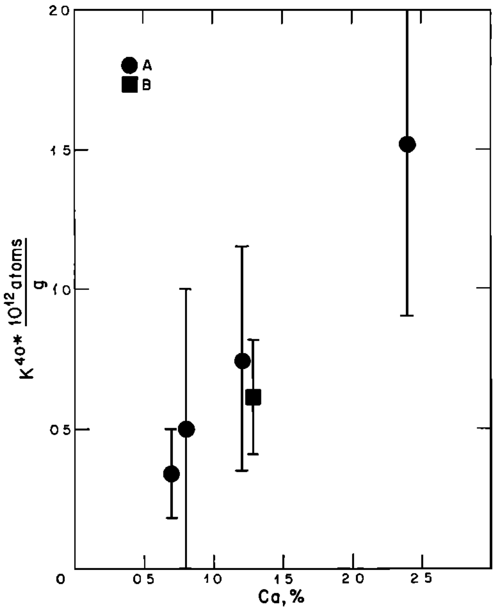

Fig. 8. Plot showing the correlation between the amount of excess K^{40}* in various samples of Norton County with the Ca concentration measured in the same sample. The circles denote samples obtained from C. Moore. The square denotes sample obtained from W. Nichiporuk.

rate of about $2.7 \pm 1.4 \times 10^{11}$ atoms K^{40}/g Ca per 10^{9} years.

According to *Prior and Hey* [1953], Pena Blanca Springs and Norton County are both aubrites, although *Beck and La Paz* [1951] have argued for a special classification for Norton County. If the K^{40} enrichments in Norton County represented something more fundamental than cosmic-ray production, and if petrological classifications really group meteorites that have a common origin, all aubrites might have been expected to show enrichments in K^{40}. However, no enrichment could be detected in Pena Blanca Springs. But, if the enrichment in Norton County is cosmogenic, the lack of it in Pena Blanca Springs is understandable because the K content is 4 times larger and the cosmic ray exposure age 4 to 5 times smaller than for Norton County [*Eberhardt et al.,* 1965].

The above discussion suggests that $Ca^{40}(n, p)$ is the source of the observed enrichments; however, there is no evidence for the $Mg^{24}(n, \alpha)Ne^{21}$ reaction which should also occur at approximately the same neutron energy. *Kirsten et al.* [1963] measured $Ne^{20}:Ne^{21}:Ne^{22} = 53:58:63 \times 10^{-8}$ cm^3 (at STP) in Norton County. Approximately equal yields of the Ne isotopes is a property of high-energy (> 50 Mev) reactions, but Ne^{21} should have been in excess if low-energy neutron reactions were important because of the high abundance of Mg^{24}. No experimental cross-section data are available for $Mg^{24}(n, \alpha)$; thus the theoretical excitation function of *Bullock and Moore* [1960] (curve *B* in Figure 9 of their paper) was used along with the AHL energy spectrum at 10 g/cm^2 depth to calculate $(Ne^{21*}/Mg^{24})/(K^{40*}/Ca^{40}) = 0.23$ for the 2- to 20-Mev energy range. The uncertainties in this ratio are mostly from the cross-section estimates because errors in the flux spectrum tend to cancel; thus it should be reliable to within a factor of 3. Using $K^{40*} = 6 \times 10^{11}$ atoms/g for 1% Ca from Figure 8 and 25% for the Mg concentration [*Wiik,* 1956], we calculated the amount of Ne^{21} produced from $Mg^{24}(n, \alpha)$ by 2- to 20-Mev neutrons to be about 17×10^{-8} cm^3 (at STP). This should have resulted in an easily measured excess of Ne^{21} over Ne^{20} and Ne^{22}, but it was not observed. It is possible that the apparent discrepancy is merely due to the uncertainties in the above estimate; nevertheless, the problem deserves further study.

Although some questions remain unanswered, the bulk of the evidence indicates that the K^{40*} enrichments in Norton County are cosmogenic and are due to the $Ca^{40}(n, p)$ reaction. In the remainder of our discussion we will assume these to be true, although we clearly have not proved either point conclusively.

Weekeroo Station and Vaca Muerta. Data for the Weekeroo Station iron and anorthite from the Vaca Muerta mesosiderite are also given in Table 5 and Figure 6.

High K concentrations were observed for both samples of Weekeroo Station silicate. This is not mineralogically unreasonable because plagioclase and diopside are the major minerals in the silicate phase [C. Frondel, private communication]. However, this meteorite is more likely to be contaminated because it was found in 1924 and its terrestrial age is unknown [*Hodge-Smith,* 1930]. Also, the inclusions are interconnected by an extensive system of small cracks, which means that a large number of the inclusions were probably connected to the original surface by these cracks. This allows the possibility of exposure to groundwater or to cooling water during cutting and polishing operations; however, visual inspection of the silicate samples gave no evidence of this. Our piece was a sawed slab which had been polished on one side and shellacked on both sides, and it is possible that some of the shellac had entered the cracks or soaked directly into the somewhat porous inclusions. For these reasons great care was taken in extracting the silicate, as discussed in section 2.

The significance of the concentration measurements is somewhat limited by the fact that no thorough magnetic separation was performed on the silicate samples; however, the difference in the K and Ca concentrations are too large to be accounted for by this limitation.

The Weekeroo I sample has about a 0.8% enrichment in K^{40}, which corresponds to about 2×10^{-9} g K^{40*}/g meteorite. The silicate samples are no more than ⅓ iron-group elements; thus there is about 6×10^{-9} g K^{40*}/g iron. This is 7 to 50 times larger than that observed by Stauffer and Honda in the iron phase of other iron meteorites. This would require 5 to 10 $\times$ 10^9 years exposure age, which is quite high. However, Weekeroo I has 5 times as much Ca as the Norton County samples and 30 times as much K^{40*}. Thus only about a factor of 6 in exposure age, or 1.2 to 1.4 $\times$ 10^9 years, is required to account for the observed enrichment using the $Ca^{40}(n, p)$ reaction. The absence of an observable enrichment in Weekeroo D can be understood because it has twice as much K and 40% less Ca than Weekeroo I.

Similarly, the 1.5% enrichment measured in the Vaca Muerta anorthite can be obtained from the $Ca^{40}(n, p)$ reaction because the anorthite has 13 times more Ca but only 6 times more K^{40*} than Norton County. This requires a cosmic-ray exposure age of only about 10^8 years. Spallation of Fe-group nuclei should produce negligible K^{40*} because these are present only as trace elements in the anorthite.

As a result of the above arguments, we assume that the K^{40} enrichments in Norton County, Vaca Muerta, and Weekeroo Station are due to cosmic-ray production by the $Ca^{40}(n, p)$ reaction and are not evidence for nucleosynthesis in the early history of the solar system. This indicates that the primordial K^{40} abundance in all of the meteorites was within 0.5 to 1% of the terrestrial value. Interpreting the enrichments in terms of a difference in the fraction of terrestrial and meteoritic material which had been irradiated would yield a maximum difference of 5%.

Summary. Table 6 presents our 'best estimates' of the $(K^{41}/K^{40})_c$ ratios for all samples. These estimates were obtained by averaging selected measurements from Tables 1–4. Criteria used in selection were stability, flatness of the base line near the K^{40} peak, and internal precision as indicated by the statistical error. An uncertainty of ± 2 has been assigned to the numbers in Table 6 on the basis of frequency distribution in $(K^{41}/K^{40})_c$ for the terrestrial samples given in Figure 4*a*.

4. Conclusion

The FGH irradiation. The results of the present measurements show that there are no measurable differences in the K^{40} abundance between terrestrial samples and the 11 meteorites examined which cannot be accounted for by cosmic-ray production. Any variations in the K^{40} abundance produced by nuclear reactions are probably less than 0.5% and certainly less than 1%. From the arguments given in section 1, we see that any variations in the fraction, q, of material irradiated between terrestrial and meteoritic material are less than 1 to 2%. Conclusions will now be drawn assuming that this result applies in particular to all meteorites and, more generally, to all the solid objects in the solar system. This latter assumption should be verified when other samples of extraterrestrial material become available.

As discussed in section 1, the interpretation of the results in terms of a limit on variations in q depends on the assumption that all samples have been exposed to the same flux, as implied by the Li^6/Li^7 measurements on meteorites by *Krankowsky and Müller* [1964]. The present experiments set limits on the product of variations in the flux and in q that are independent of the Krankowsky-Müller experiments. The 1 to 2% limit set on q is approximate because it depends on the rather uncertain parameters used in the FGH calculation [*Burnett et al.*, 1965*a*]. The importance of the $Ca^{40}(n, p)$ reaction in producing cosmogenic K^{40} suggests that neglecting this reaction caused the calculated amount of K^{40} synthesized in the FGH process to be underestimated, which means that still lower limits might be set on variations in q.

We have thus failed to find any direct evidence for the occurrence of a large-scale irradiation in the early history of the solar system. Variations in the K^{40} abundance would have been expected on the basis of the usually accepted assumption that meteorites come from the asteroid belt and are formed there or in some other region of space distant from the earth. On the other hand, if a large-scale irradiation did take place, the present results set limitations on possible mechanisms for the formation of the earth and the meteorite parent bodies. A more detailed discussion of particular mechanisms is given by *Burnett et al.* [1965*a*]; however, in general terms the conclusion is that (*a*) the flux and size distribution of the planetesimals were constant throughout the inner solar system, (*b*) the flux varied but all the material was concentrated in one region of space, or (*c*) the material was widely distributed but thoroughly mixed during or after the irradiation. For alternatives (*b*) and (*c*) we can also conclude that the process which caused the present-day separation of material in the inner solar system deposited planetesimals with the same size distribution—or, more generally, the same relative amounts of irradiated and unirradiated material—in each location. That part of the general conclusion based on a constant flux may be drawn from the Krankowsky-Müller experiment alone. The total conclusion may be drawn from the present experiment alone, although less precisely.

The above conclusions do not eliminate the possibility of fine-grained or microscopic isotopic inhomogeneities because they are based on isotopic analyses of 0.1- to 1-g samples. Some isotopic data on chondrules have been reported, however. *Murthy and Sandoval* [1965] find no variation in Cr^{54} to within 1% in chondrules from four meteorites and G. Wetherill (private communication, 1965) finds no measurable variation in K^{40} for chondrules from Bruderheim. Thus it would appear that the irradiation history of these bodies is not different from that of the bulk of meteoritic and terrestrial matter.

The measurement of the isotopic composition of Gd, Sm, and Eu by *Murthy and Schmitt* [1963] and of Li by *Krankowsky and Müller* [1964] were only for stone meteorites. *Urey* [1965] and *Arnold* [1965] have pointed out that a lunar origin for those stone meteorites which have short cosmic-ray exposure ages cannot be disproved. Thus, in the absence of any data on these key elements for irons, a lunar origin for the stones could conceivably explain the apparently identical irradiation and mixing history of the earth and the stones. But, according to Arnold, the high exposure ages of irons eliminate a lunar origin and thus indicate an asteroidal origin. The amount of cosmogenic K^{40} observed in Weekeroo Station silicate is consistent with a high exposure age and, from the above analysis, could come from the asteroid belt. The terrestrial K^{40} abundance found for Weekeroo Station silicate implies that the difficulties for the FGH model would remain even if stone meteorites have a lunar origin. A more detailed discussion of this point is given by *Burnett et al.* [1965*a*].

Murthy and Sandoval [1965] found no measurable differences to within 1% in the isotopic abundance of Cr^{54} among various meteorites and reagent Cr. Cr^{54} has a high yield in the spallation of iron, and large amounts would be produced by the FGH irradiation. *Hulston and Thode* [1965] find no variations in S^{33} that cannot be explained by chemical fractionation processes. Deviations of 0.1% due to nuclear effects could probably have been detected. The amount of S^{33} would be significantly increased in the irradiated material by neutron capture on S^{32} in the FGH process. It should be noted that both sets of authors discuss their results in terms of a comparison between pure irradiated and pure unirradiated material, assuming that only partial mixing has occurred.

Based primarily on the experiments of Murthy and Schmitt, our model requires that the irradiated and unirradiated portions of both terrestrial and meteoritic material have been thoroughly mixed. Interpreting the Cr^{54} and S^{33} results on this basis, as was done for K^{40} in section 1, we estimate that the 1% limits on Cr^{54} set about 20% limits and the 0.1% limits on S^{33} set about 6% limits on variations in q.

Clayton [1963] measured an unusually high enrichment of 6% of C^{13} from the carbonate minerals in Orgueil and suggested that this may have been nuclear in origin and possibly due to the FGH irradiation. The FGH process is able to synthesize all the terrestrial C^{13} [*Burnett et al.*, 1965*a*]; thus the 6% C^{13} enrichment can be explained if the material of Orgueil experienced a 6% larger flux or has 6% more irradiated material. In either case, easily measurable effects in K^{40} would have resulted which are not present. This suggests that the C^{13} enrichment is geochemical. Alternatively, a different mixing ratio could be proposed for meteoritic and terrestrial C and the same for meteoritic and terrestrial K, but this seems unreasonable.

Limits on differential uniform irradiations. The low abundance of K^{40} compared with neighboring nuclides, e.g., K^{39}, Ca^{40}, and Fe^{56}, means that it is a sensitive indicator for particle irradiations because only a very small fraction of the target nuclei need react in order to produce a measurable effect on the K^{40} abundance. This sensitivity allows us to investigate primordial fluxes which are much less than those postulated by FGH. The K^{40} measurements can be used to set limits on *differences* in the integrated particle flux between terrestrial and meteoritic material for the time when they were sufficiently widely dispersed so that all the material was irradiated. The measurements by *Voshage and Hintenberger* [1963] of cosmogenic K in iron meteorites show that there have been no significant irradiations greater than a factor of 2 larger than that due to the cosmic rays in the last 10^9 years.

Limits can be set for high-energy particles, thermal neutrons, and 2- to 20-Mev neutrons. For high-energy particles, assuming 8 mb as an effective spallation cross section on the basis of measurements of *Honda and Lal* [1964], limits of 1 to 10 $\times$ 10^{17} particles/cm^2 corresponding to less than 1% enrichments in K^{40} can be set for the meteorites we have studied, depending on their Fe and K concentration. This corresponds to about 1 to 5 $\times$ 10^9 years of cosmic-ray bombardment. For the special case of the carbonaceous chondrites the limit is about 3 $\times$ 10^{17} particles/cm^2, assuming 400 ppm K and 25% Fe. *Krummenacher et al.* [1962] measured about 3 $\times$ 10^9 atoms/g of excess Xe^{124} in Orgueil and Murray using either Xe^{132} or Xe^{130} as the reference isotope. In situ spallation has been considered as a likely source of the excess Xe^{124} (see, for example, *Anders* [1964]). If an average target concentration of 5 ppm at about mass 140 and an effective spallation cross section of 10 mb are assumed, a high-energy flux of about 10^{19} particles/cm^2 is required. This flux would have caused a 3% effect in K^{40}, but it was not observed. Considering the uncertainty in the above calculation, it cannot definitely be said that a discrepancy exists; however, it does raise a problem that requires more study.

A 1% enrichment in K^{40} requires about 5 $\times$ 10^{17} thermal neutrons/cm^2 regardless of the K concentration. This calculation applies to uniform irradiation and not to the FGH process, which involves mixing of irradiated and unirradiated material. A considerably smaller upper limit of 4 $\times$ 10^{16} neutrons/cm^2 can be set based on the Gd^{157} abundance measurement by *Murthy and Schmitt* [1963].

Assuming the shape of the cosmic-ray energy spectrum in the range 2 to 20 Mev given by *Arnold et al.*, [1961] and the excitation function assumed in section 3, we find that the average cross section for the $Ca^{40}(n, p)$ reaction is about 400 mb. For a sample with Ca/K = 10 an integrated flux of 3 $\times$ 10^{17} neutrons/cm^2 is required to produce a 1% effect in the K^{40} abundance.

The sensitivity of K^{40} and Gd^{157} to nuclear reactions means that measurements of their abundances would be significant in establishing the irradiation history of the lunar surface and other extraterrestrial samples. Even if particle fluxes larger than present-day cosmic rays have never occurred, observable effects should be present.

Table 7 summarizes our estimates of the limits set on differences in primordial nuclear irradiations in the early history of the solar system by various isotopic measurements on terrestrial and meteoritic samples. This has been done for both the FGH model and a uniform irradiation model. The limits on the integrated particle flux for uniform irradiation are clearly order of magnitude. The important point is the order of magnitude and relative sensitivities of various stable nuclei.

K-Ar ages of meteorites. In general the present measurements have verified the assumption of a terrestrial isotopic composition for

TABLE 7. Limits on Differential Irradiation History of Earth and Meteorites by Isotopic Measurements

Nuclide(s)	Uncertainty in Experimental Abundances, %	FGH Model		Uniform Irradiation[a]	
		Parameter	Limits on Allowed Variations, %	Energy Range of Particle Flux	Integrated Flux Limits, particles/cm²
Li^7/Li^6 [b]	2	Flux	≈2	≲ 75 Mev	1×10^{18}
Gd^{157} [c]	1	q	20	Thermal neutrons	4×10^{16}
S^{33} [d]	0.1	q	6	Thermal neutrons	1×10^{19}
S^{36} [d]	0.1			>1000 Mev	10^{20}
Cr^{54} [e]	1	q	20	> 30 Mev	3×10^{19}
K^{40} [f]	0.5–1	q	1–2	> 500 Mev	2×10^{17}
				Thermal neutrons	5×10^{17}
				2–20 Mev neutrons	3×10^{17}
V^{50}	2			> 100 Mev	5×10^{17}

[a] Assuming type I carbonaceous chondrite chemical composition.
[b] *Krankowsky and Müller* [1964].
[c] *Murthy and Schmitt* [1963].
[d] *Hulston and Thode* [1965].
[e] *Murthy and Sandoval* [1965].
[f] Present work.

meteoritic K which is always used in K-Ar dating of both stone and iron meteorites.

The large concentration of K found in the Weekeroo Station silicate enables us to show that the primordial K in at least this iron meteorite has the terrestrial K^{40} abundance within 1% because the corrections for cosmogenic K^{40} were small and that for K^{41} negligible. This is of interest because *Stoenner and Zähringer* [1958] and *Fisher* [1965], using the method of *Zähringer and Fireman* [1956], measured Ar^{40}/K^{41} ratios which would imply ages of 8 to 13 × 10^9 years if the terrestrial K^{41}/K^{40} ratio is assumed. This is much higher than the accepted 4.5 × 10^9 years for the formation of the earth and the meteorites [*Patterson,* 1956]. Fisher has shown that corrections for cosmogenic Ar^{40}, K^{40}, and K^{41} do not resolve the problem, and he also gives indirect arguments to show that the K^{40} abundance in iron meteorites is terrestrial. Our measurement of Weekeroo Station, to the extent that it is representative of all iron meteorites, demonstrates this and eliminates it as an explanation of the high apparent K-Ar ages of iron meteorites.

Let us assume that the results for the Weekeroo Station silicate imply that the primordial K^{40} abundance was terrestrial in the iron phase of all iron meteorites. Then, if the iron meteorites did form 10^{10} years ago, the remainder of the solar system must have been isolated from an event of nucleosynthesis for the same length of time; because, owing to the short half-life of K^{40} compared with 5.5 × 10^9 years, only small amounts of new elements need be added to produce an observable variation between the irons on one hand and the stone meteorites and the earth on the other. This conclusion can be inferred more decisively from K^{40} measurements than from isotopic measurements of stable elements because it is true even if the isotopic compositions of the elements have been the same during the entire history of the galaxy. Thus it does not seem possible that iron meteorites condensed before the formation of the solar system, as suggested by *Marshall* [1962], assuming nucleosynthesis continued in the intervening time.

Similar conclusions can be drawn from a measurement of the Rb^{85}/Rb^{87} ratio, although less decisively because of the long half-life of Rb^{87}. A measurement of this ratio for Weekeroo Station gave 2.63; whereas a terrestrial sample of reagent Rb measured as a standard gave 2.60. Rb analyses in this laboratory typically give ratios in the range 2.60 to 2.62. Any variation in Rb^{85}/Rb^{87} must therefore be less than 2%.

The required period of isolation raises some problems: (1) the initial U^{235}/U^{238} ratio would have to be 27, which seems unreasonably high. (2) The grouping of Pb-Pb ages [*Patterson*, 1956] with the Rb-Sr ages [*Gast*, 1962; *Pinson et al.*, 1965; *Murthy and Compston*, 1965] and many K-Ar ages (see, for example, *Kirsten et al.* [1963]) would be hard to understand because the primordial Pb^{207}/Pb^{206} ratio is based on the Canyon Diablo iron meteorite. K-Ar and particularly Rb-Sr ages of silicate inclusions in iron meteorites would be of great importance. Work on this problem is currently in progress in this laboratory. (3) The proposed existence of I^{129} [*Reynolds*, 1963] and Pu^{244} [*Rowe and Kuroda*, 1965; *Fleischer et al.*, 1965] at the time of formation of the solar system would be hard to understand because of their short half-lives. Although nucleosynthesis in the solar nebula might account for I^{129}, this does not seem possible for Pu^{244}.

Several of the meteorites investigated in this study were chosen on the basis of the high Ar^{40}-K^{40} ages obtained for them by other workers [see *Kirsten et al.*, 1963]. Of these only Norton County contained an enrichment of K^{40}, and Ar^{40}-K^{40} ages for it ranging from 2.3 to 5.09×10^9 years, have been reported by *Geiss and Hess* [1958], *Vinogradov et al.* [1960], and *Kirsten et al.* [1963].

The possibility that the age of 5.09×10^9 years reported by Kirsten et al. is real requires careful consideration. The Ar^{40}/K^{40} ratio is about 41% higher than would be obtained for an age of 4.5×10^9 years. Our data show that there are no anomalies in Norton County sufficient to produce the Ar^{40} by radioactive decay. The direct production of Ar^{40} by spallation could, in principle, have a significant effect on the age; however, the contents of cosmogenic Ar and Ne found in this meteorite by all workers is far below the level necessary to account for this. The ratio of Ar^{40}/Ar^{36} is around 400 and is therefore subject to large error if there is any atmospheric or primordial contamination present. However, the amount of atmospheric Ar^{36} which would accompany the Ar^{40} would be 0.67×10^{-8} cm^3/g (at STP). Such an explanation of this anomaly would require a ratio of (Ar^{38}/Ar^{36}) from spallation of 2.4, which is higher than the value of 1.4 to 1.6 indicated by other data for spallation of Fe-group nuclei (see, for example, *Begemann* [1965]). Spallation on Ca, which is so abundant in this meteorite in comparison with the Fe-group elements, might possibly give this high Ar^{38}/Ar^{36} ratio.

With the exception of the data by *Vinogradov et al.* [1960], all the K contents reported are about 70 ppm. The present data indicate that the K content may range over a factor of 7 and show that the problem of sampling for Norton County may be very difficult for Ar and K analyses on different parts of the same sample.

From the present data it appears difficult to resolve this anomalous age, as well as the Ne^{21} problem discussed in section 3. If the greater age is correct, it may be possible to observe very large effects in Xe^{129} from I^{129} decay. This meteorite requires more study.

Summary of conclusions. The present measurements show that there are no variations in the abundance of K^{40} due to nuclear processes to within 0.5 to 1%, and at present it appears that the isotopic abundances of all elements between terrestrial and meteoritic material show no variations that can be attributed to a large-scale particle irradiation in the early history of the solar system. However, an important exception may be Xe; variations in the isotopic composition of meteoritic Xe exist for which no generally accepted explanation has been proposed. In the present context the variations in Xe^{124}, Xe^{126}, and Xe^{128}, which cannot be radiogenic, appear to be most significant because they can be produced in significant quantities by spallation reactions. The sensitive elements, for the FGH model, Li, Gd, S, and K, have now been examined, and further investigations of this kind appear fruitless with respect to investigating the validity of the FGH process. If the FGH model is to be retained, a plausible astrophysical process must be postulated for the formation of the earth and the meteorites which will explain the observed isotopic homogeneity.

Upper limits for differences in primordial integrated particle fluxes for uniform irradiation conditions have been set for irradiation by high-energy particles, thermal neutrons, and 2- to 20-Mev neutrons. Further investigation seems necessary to determine whether the upper limit for the high-energy flux of 2×10^{17} particles/cm^2 can satisfactorily account for the amounts of Xe^{124} observed in the carbonaceous chondrites.

The assumption of a terrestrial K^{40} isotopic abundance for calculating K-Ar ages for both stone and iron meteorites has been confirmed. The lack of significant K^{40} variations in the silicate inclusions from Weekeroo Station shows that the iron meteorites cannot be older than the solar system. The large amounts of K found in Weekeroo Station inclusions means that unambiguous K-Ar and Rb-Sr age measurements are possible for this and other iron meteorites.

Acknowledgments. We wish to thank Professor R. E. Folinsbee, Professor C. Frondel, Walter Nichiporuk, and Professor L. T. Silver for meteorite samples used in this work. We wish to acknowledge especially the cooperation of Professor C. B. Moore and the Nininger Meteorite Collection in obtaining and discussing meteorite samples. We are grateful to Professors W. A. Fowler and L. T. Silver for valuable discussions.

This work was supported in part by the Office of Naval Research [Nonr-220(47)], the National Aeronautics and Space Administration [NGR-05-002-028], the U. S. Atomic Energy Commission [AT(04-3)-427], and the National Science Foundation.

REFERENCES

Allen, L., Jr., W. A. Biggers, R. J. Prestwood, and R. K. Smith, Cross sections for the $S^{32}(n, p)P^{32}$ and the $S^{34}(n, \alpha)Si^{31}$ reactions, *Phys. Rev., 107,* 1363, 1957.

Anders, E., Meteorite ages, in *The Moon, Meteorites and Comets,* vol. 4 of *The Solar System,* edited by B. M. Middlehurst and G. P. Kuiper, University of Chicago Press, 1963.

Anders, E., Origin, age, and composition of meteorites, *Space Sci. Rev., 3,* 583–714, 1964.

Arnold, J. R., The origin of meteorites as small bodies, 2 and 3, *Astrophys. J., 141,* 1536 and 1548, 1965.

Arnold, J. R., M. Honda, and D. Lal, Record of cosmic-ray intensity in the meteorites, *J. Geophys. Res., 66,* 3519–3531, 1961.

Beck, C. W., and L. La Paz, The nortonite fall and its mineralogy, *Am. Mineralogist, 36,* 45, 1951.

Begemann, F., Edelgasmessungen an Eisenmeteoriten und deren Einschlussen, *Z. Naturforsch., 20a,* 950, 1965.

Begemann, F., J. Geiss, and D. C. Hess, Radiation age of a meteorite from cosmic-ray-produced He^3 and H^3, *Phys. Rev., 107,* 540–542, 1957.

Begemann, F., and E. Vilcsek, Durch Spallationsreaktionen und Neutroneneinfang erzeugtes Cl^{36} in Meteoriten und die Prae-Atmosphärische grosse von Steinmeteoriten, *Z. Naturforsch., 20a,* 533, 1965.

Bullock, R. E., and R. G. Moore, Jr., Odd-even dependence of nuclear level density parameters, *Phys. Rev., 119,* 721–731, 1960.

Burnett, D. S., W. A. Fowler, and F. Hoyle, Nucleosynthesis in the early history of the solar system, *Geochim. Cosmochim. Acta,* in press, 1965*a*.

Burnett, D. S., H. J. Lippolt, and G. T. Wasserburg, The isotopic abundance of K^{40} in stone meteorites (abstract), *Trans. Am. Geophys. Union, 46,* 125, 1965*b*.

Clayton, R. N., Carbonate isotope abundance in meteoritic carbonates, *Science, 140,* 192–193, 1963.

Eberhardt, P., O. Eugster, and J. Geiss, Radiation ages of aubrites, *J. Geophys. Res., 70,* 4427–4434, 1965.

Edwards, G., and H. C. Urey, Determination of alkali metals in meteorites by a distillation process, *Geochim. Cosmochim. Acta, 7,* 154–168, 1955.

Fireman, E. L., and J. De Felice, Argon-39 and tritium in meteorites, *Geochim. Cosmochim. Acta, 18,* 183–192, 1960.

Fisher, D. E., Anomalous Ar^{40} contents in iron meteorites, *J. Geophys. Res., 70,* 2445–2452, 1965.

Fleischer, R. L., P. B. Price, and R. M. Walker, Spontaneous fission tracks from extinct Pu^{244} in meteorites and the early history of the solar system, *J. Geophys. Res., 70,* 2703–2707, 1965.

Fowler, W. A., J. L. Greenstein, and F. Hoyle, Nucleosynthesis during the early history of the solar system, *Geophys. J., 6,* 148–220, 1962.

Gast, P. W., The isotopic composition of strontium and the age of stone meteorites, 1, *Geochim. Cosmochim. Acta, 26,* 927–943, 1962.

Gast, P. W., Terrestrial ratio of potassium to rubidium and the composition of the earth's mantle, *Science, 147,* 858–860, 1965.

Geiss, J., and D. C. Hess, Argon-potassium ages and the isotopic composition of argon from meteorites, *Astrophys. J., 127,* 224–236, 1958.

Hess, W. N., H. W. Patterson, R. Wallace, and E. L. Chupp, Cosmic-ray neutron energy spectrum, *Phys. Rev., 116,* 445, 1959.

Hirt, B., and S. Epstein, A search for isotopic variations in some terrestrial and meteoritic calcium (abstract), *Trans. Am. Geophys. Union, 45,* 113, 1964.

Hodge-Smith, T., The Weekeroo meteorite: A siderite from South Australia, *Records Australian Museum, 18,* 312, 1930.

Honda, M., and D. Lal, Spallation cross sections for long-lived radionuclides in iron and light nuclei, *Nucl. Phys., 51,* 363–368, 1964.

Honda, M., S. Umemoto, and J. R. Arnold, Radioactive species produced by cosmic rays in Bruderheim and other stone meteorites, *J. Geophys. Res., 66,* 3541–3546, 1961.

Hulston, J. R., and H. G. Thode, Variations in the S^{33}, S^{34}, and S^{36} contents of meteorites and their relation to chemical and nuclear effects, *J. Geophys. Res., 70,* 3475–3484, 1965.

Kaviladze, M. S., and I. V. Abashidze, On the variation of the isotopic ratio K^{39}/K^{41} in the earth's crust, *Bull. Acad. Sci. Georgian SSR, 35,* 67, 1964.

Keil, K., and K. Fredriksson, Electron microprobe analysis of some rare minerals in the Norton County achondrite, *Geochim. Cosmochim Acta, 27,* 939–947, 1963.

Kendall, B. R. F., Isotopic composition of potassium, *Nature,* 186, 225–226, 1960, and Ph.D. thesis, University of Western Australia, 1960.

Kirsten, T., D. Krankowsky, and J. Zähringer, Edelgas-und Kalium-Bestimmungen an einer grösseren Zahl von Steinmeteoriten, *Geochim. Cosmochim. Acta, 27,* 13–42, 1963.

Krankowsky, D., and O. Müller, Isotopenhäufigkeit und Konzentration des Lithiums in Steinmeteoriten, *Geochim. Cosmochim. Acta, 28,* 1625–1630, 1964.

Krummenacher, D., C. M. Merrihue, R. O. Pepin, and J. H. Reynolds, Meteoritic krypton and barium versus the general isotopic anomalies in meteoritic xenon, *Geochim. Cosmochim. Acta, 26,* 231–250, 1962.

Létolle, R., Sur l'abondance relative de l'isotope 41 du potassium suivant son origine géologique, *Compt. Rend., 257,* 3996, 1963.

Marshall, R. R., Cosmic radiation and the K^{40}-Ar^{40} "ages" of iron meteorites, *Geochim. Cosmochim. Acta, 26,* 983–994, 1962.

Mullins, L., and K. Zerahn, The distribution of potassium isotopes in biological material, *J. Biol. Chem., 174,* 107, 1948.

Murthy, V. R., and W. Compston, Rb-Sr ages of chondrules and carbonaceous chondrites, *J. Geophys. Res., 70,* 5297–5307, 1965.

Murthy, V. R., and P. Sandoval, Chromium isotopes in meteorites, *J. Geophys. Res., 70,* 4379–4382, 1965.

Murthy, V. R., and R. A. Schmitt, Isotope abundances of rare-earth elements in meteorites, 1, Implications of samarium, europium, and gadolinium to the early history of the solar system, *J. Geophys. Res., 68,* 911–917, 1963.

Nier, A. O., A redetermination of the relative abundances of the isotopes of carbon, nitrogen, oxygen, argon, and potassium, *Phys. Rev., 77,* 789, 1950.

Patterson, C. C., Age of meteorites and the earth, *Geochim. Cosmochim. Acta, 10,* 230–237, 1956.

Pinson, W. H., Jr., C. C. Schnetzler, E. Beiser, H. W. Fairbairn, and P. M. Hurley, Rb-Sr age of stony meteorites, *Geochim. Cosmochim. Acta, 29,* 455–466, 1965.

Prior, G. T., and M. H. Hey, *Catalogue of Meteorites,* William Clowes and Sons, London, 1953.

Reuterswärd, C., On the isotopic composition of potassium, *Arkiv Fysik, 11,* 1, 1956.

Reynolds, J. H., Xenology, *J. Geophys. Res., 68,* 2939–2956, 1963.

Rik, G. R., and I. A. Shukolyukov, Composition of K in meteorites, *Dokl. Akad. Nauk SSSR, 94,* 667, 1954.

Rowe, M. W., and P. K. Kuroda, Fissiogenic xenon from the Pasamonte meteorite, *J. Geophys. Res., 70,* 709–714, 1965.

Schumb, W. C., R. D. Evans, and W. M. Leaders, Radioactive determination of the relative abundance of the isotope K^{40} in terrestrial and meteoritic potassium, *J. Am. Chem. Soc., 63,* 1203, 1941.

Stauffer, H., and M. Honda, Cosmic-ray-produced stable isotopes in iron meteorites, *J. Geophys. Res., 67,* 3503–3512, 1962.

Stoenner, R. W., and J. Zähringer, Potassium-argon age of iron meteorites, *Geochim. Cosmochim. Acta, 15,* 40–50, 1958.

Suess, H. E., On the age of the elements, *Naturwiss., 26,* 411, 1938.

Urech, S., E. Jennet, and J. Rossel, Les réactions (n, p) et (n, α) de Ca^{40} avec des neutrons de 6 meV, *Helv. Phys. Acta, 34,* 954, 1961.

Urey, H. C., Meteorites and the moon, *Science, 147,* 1262, 1965.

Vinogradov, A. P., I. K. Sadoroschnii, and K. G. Knorre, Argon in meteorites, *Meteoritika, 18,* 92–99, 1960.

Voshage, H., and H. Hintenberger, The cosmic ray exposure ages of iron meteorites as derived from the isotopic composition of potassium and the production rates of cosmogenic nuclides in the past, in *Radioactive Dating,* pp. 367–379, IAEA, Vienna, 1963.

Wasserburg, G. J., T. Wen, and J. Aronson, Strontium contamination in mineral analyses, *Geochim. Cosmochim. Acta, 28,* 407–410, 1964.

Wiik, H. B., The chemical composition of some stony meteorites, *Geochim. Cosmochim. Acta, 9,* 279–289, 1956.

Zähringer, J., and E. L. Fireman, The Ar^{40}, K^{41} and He^{3} content of iron meteorites (abstract), *Bull. Am. Phys. Soc., 1,* 344, 1956.

(Manuscript received October 15, 1965.)