Proc. Lunar Sci. Conf. 6th (1975), p. 1407–1418.
Printed in the United States of America

Primordial and cosmogenic radionuclides in Descartes and Taurus-Littrow materials: Extension of studies by nondestructive γ-ray spectrometry*

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Abstract—Our previous studies on the distributions of K, Th, U, ^{22}Na, ^{26}Al, and ^{54}Mn in material from Apollos 16 and 17 have been extended to include 12 samples from additional sampling stations and with a greater range of properties. Samples were: from Descartes, 60618, 63320, 63340, 65785, 67016, 67115; and from Taurus-Littrow, 70315, 70321, 71546, 72155, 72161, 74245.

Soils from Station 13 at Descartes were 20–30% lower in primordial radioelements than for average amounts at eight other stations. Two light-matrix breccias, 67016 and 67115, contained 3–5 times smaller concentrations of K, Th, and U than those found for other breccias at North Ray Crater. Anorthositic rocks 60618 and 65785 have K/U ratios near the average values for Apollo 16 materials, but 60618 exhibits a Th/U ratio of only 2.3, the lowest we have observed for Apollo 16 materials.

Surface soil 72161 was collected in a dark valley floor area adjacent to the light-mantling detritus from the South Massif, but its primordial radioelement content most closely resembles that of North Massif soils. The distribution of K, Th, and U in Apollo 17 basalts supports the trends we observed earlier. Values for Th/U of 2.7 ± 0.3 and K/U of 4200 ± 500 are typical of coarse and medium basalts. The fine basalts generally have Th/U > 3.0 and K/U of 3000 ± 500, which supports the speculation that two or three subfloor basalt units were sampled at Taurus-Littrow.

Cosmogenic radionuclide contents of shadowed soil 63320 indicate that the shielding geometry of Shadow Rock was established about 3 m.y. ago and that the recent shielding of 63320 from solar cosmic rays was only $\sim 80\%$. The following rocks were found to have concentrations of ^{26}Al below the expected saturation level, indicating a short exposure or partial shielding: 67016, 67115, 67785, 70315, 71546, 72155, and 74245. The surface samples from Apollo 17 again showed the effects of the intense, proton-accelerating flare of August, 1972, particularly in the very high concentrations of ^{22}Na and ^{54}Mn.

INTRODUCTION

PREVIOUS STUDIES by Eldridge *et al.* (1973a, 1974) and O'Kelley *et al.* (1974) on suites of twenty-six samples each from the Descartes and Taurus-Littrow landing sites were performed in order to expand our knowledge of the distribution of the primordial radioelements K, Th, and U over the lunar surface. The overall concentration and distribution of these radioelements plays an important role in explanations of the moon's thermal history and its contemporary, experimentally determined heat flow.

Elemental correlations have served as indicators of differentiation among various lunar materials returned to earth by the six Apollo and the two Luna missions. The constancy of Th/U at ~ 3.8 in samples from the early Apollo

*Research carried out under Union Carbide's contract with the U.S. Atomic Energy Commission through interagency agreements with the National Aeronautics and Space Administration.

missions was noteworthy (O'Kelley *et al.*, 1970b). In samples from later Apollo missions, especially the Apollo 17 basalts, ratios of Th to U were sometimes < 3.0 (Eldridge *et al.*, 1974). Indications that separate basalt flows existed in the subfloor of Taurus-Littrow could be inferred from the correlation of Th/U and K/U ratios with sample location.

Cosmogenic radionuclide concentrations are determined simultaneously with the primordial radioelements by the technique of nondestructive γ-ray spectrometry. Cosmogenic radionuclides are those produced by an activation process whereby solar or galactic cosmic rays interact with a stable target element, producing one or more radioactive species. Comparison of concentrations of selected radionuclide pairs such as ^{26}Al and ^{22}Na may be used to calculate surface exposure ages (or apparent burial) over a time span of a few hundred thousand to a few million years. Such radionuclide determinations have also been useful in determining lunar sample orientation (O'Kelley *et al.*, 1970b) and in characterizing solar-flare activity (O'Kelley *et al.*, 1974).

EXPERIMENTAL METHODS

Data collection, system calibrations, and computer resolution of the γ-ray spectra obtained with our anti-coincidence shielded, low-level spectrometer have been previously described (O'Kelley *et al.*, 1970b) and (Eldridge *et al.*, 1973b). Calibration replicas were used for all samples in this study, and standard spectral libraries were acquired under the same experimental conditions as the lunar samples. Error values quoted include estimates of all errors, including system calibrations and counting statistics.

RESULTS AND DISCUSSION

Primordial radioelements—Apollo 16

Identification numbers of the four rocks and two soils comprising the present suite of Descartes samples are underlined in the Apollo 16 traverse map of Fig. 1. Two unsieved soil samples (63320 and 63340) collected beneath the overhang of Shadow Rock at Station 13 were measured in this study. These were originally called "permanently shadowed samples," but the geometry of the photographs was inadequate to determine whether the deep niche sampled was exposed to part of the late afternoon sun. It is likely that the sample area is exposed to direct sunlight for part of each lunar day (ALGIT, 1972). Cosmogenic radionuclide determinations discussed below support this conjecture. Shadow Rock is located on the southeast part of the North Ray Crater ejecta blanket, approximately 550 m from the crest of the crater rim. The ejecta material is fine grained; sample 63320 was taken as the representative shadowed soil, with 63340 a control sample from below 63320.

The primordial radioelement concentrations measured in the shadowed soils are shown in Table 1. It can be seen that the two soils 63320 and 63340 are identical to one another in radioelement concentrations and are slightly lower in Th and U than soil 63501 collected 8 m away. All three soils at Station 13 were

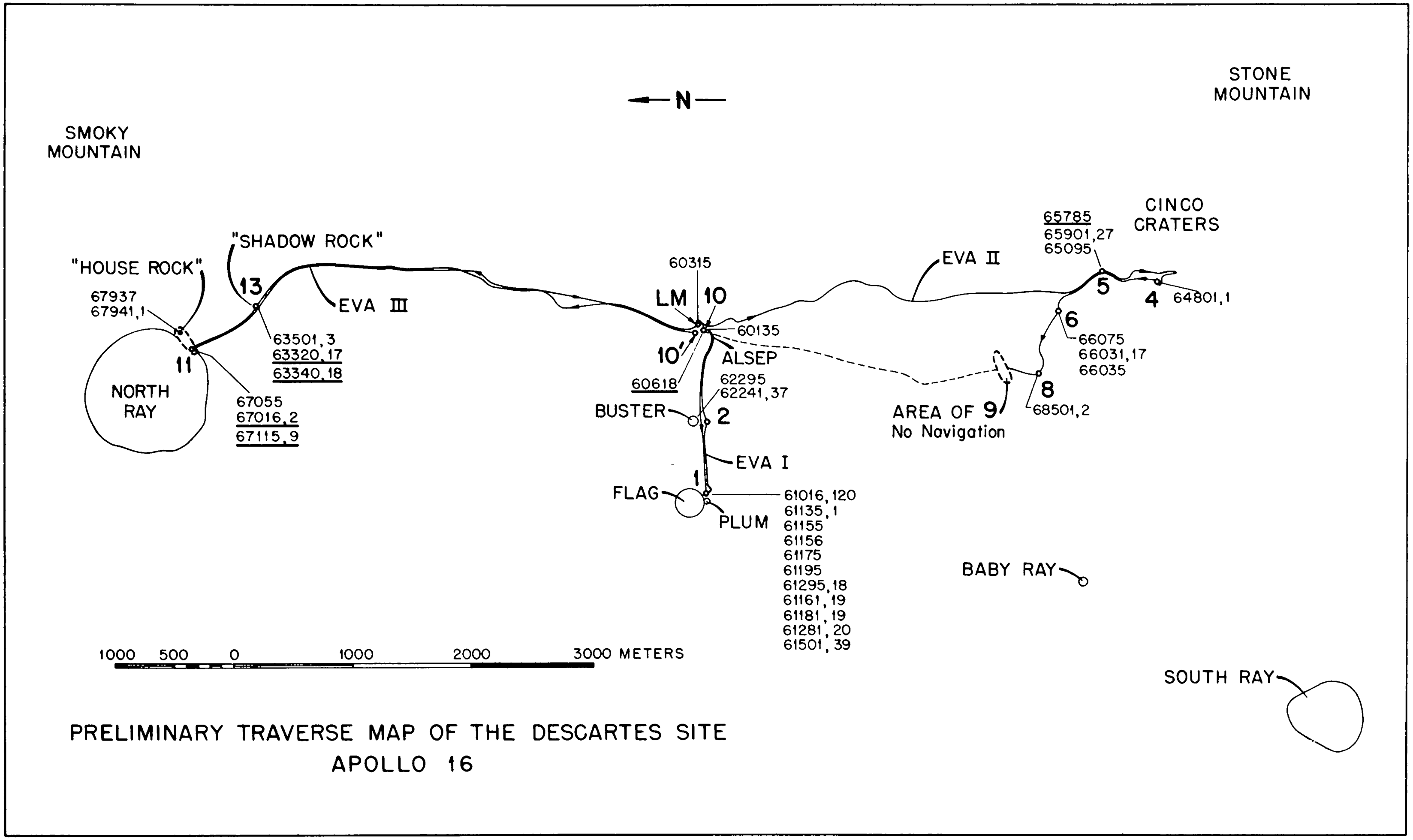

Fig. 1. Traverse map of the Apollo 16 landing site. Sampling stations are shown as large, bold-faced numerals. Underlined sample numbers are those analyzed in this study; data on other samples listed were reported by Eldridge *et al.* (1973).

Table 1. Apollo 16 radioelement concentrations.

Sample number	Type	K(ppm)	Th(ppm)	U(ppm)	Th/U	K/U
8-sta. avg.*	<1-mm fines	940 ± 50	1.95 ± .09	0.54 ± .03	3.61 ± .26	1740 ± 130
63501*	<1-mm fines	728 ± 50	1.53 ± .08	0.41 ± .03	3.73 ± .34	1780 ± 180
63320	Unsieved fines	800 ± 40	1.35 ± .07	0.39 ± .03	3.46 ± .32	2050 ± 190
63340	Unsieved fines	790 ± 40	1.33 ± .07	0.40 ± .03	3.33 ± .30	1980 ± 180
67016,2	Breccia	485 ± 25	0.69 ± .03	0.20 ± .01	3.45 ± .23	2420 ± 180
67115,9	Breccia	475 ± 25	0.44 ± .02	0.12 ± .01	3.67 ± .35	3960 ± 390
67055*	Breccia	1620 ± 80	3.69 ± .18	0.98 ± .05	3.77 ± .27	1650 ± 120
67937*	Breccia	1650 ± 90	3.24 ± .16	0.96 ± .05	3.38 ± .24	1720 ± 130
60618,0	Crystalline	670 ± 50	0.63 ± .06	0.28 ± .03	2.25 ± .32	2390 ± 310
65785,0	Crystalline	1850 ± 150	3.03 ± .18	0.97 ± .07	3.12 ± .29	1910 ± 210
62295,0*	Crystalline	630 ± 30	3.20 ± .15	0.82 ± .05	3.90 ± .30	770 ± 60

*Data previously described in Eldridge *et al.* (1973).

~20–30% lower in radioelement content than the average of 11 soils from eight stations (compare with the eight-station average listed as the first entry in Table 1).

Ulrich (1973) provided a geologic model and a stratigraphic sequence for North Ray Crater. The interpretation was based on combined evidence of rock distributions on the crater rim and photography of the crater wall. Ulrich (1973) showed that "Outhouse Rock" (67937) probably came from below 100-m depth in North Ray Crater. In our suite of samples, two breccias, 67016 and 67115, were taken at North Ray Crater. Sample 67115 was collected inside the rim crest in the vicinity of 67055 from our previous suite. Sample 67016 was collected from the rim crest. Both samples are light-matrix breccias, and radioelement concentrations for them are shown in Table 1 and are compared with 67055 and 67937 from our previous study. If stratigraphic units at the North Ray site contain similar radioelement concentrations, then 67055 came from below 100-m depth (similar to 67937) and 67016 and 67115 probably same from a shallower stratigraphic unit. Figure 2 shows a map of the Station 11 sampling area where the location of the two light-matrix breccias of this study (67016 and 67115) may be seen in relation to the previously studied breccias (67937 and 67055) that contain K, Th, and U concentrations about 3–5 times greater than 67016 and 67115. These limited sample observations would tend to verify the stratigraphic interpretation of Ulrich (1973) that the North Ray Crater event penetrated a thin Cayley layer that possibly originated from Orientale. The sub-Cayley materials from lower regions of North Ray Crater are KREEP-rich, but considerably less so than the Fra Mauro breccias we studied from the Apollo 14 collection (Eldridge *et al.*, 1972).

Two rare anorthositic rocks from the Apollo 16 rake collection were analyzed: 65785, a spinel troctolite, and 60618, a melt rock. Taylor *et al.* (1973) described the spinel troctolites as rocks consisting predominantly of plagioclase and olivine, with (Mg, Al)-spinel as a common accessory mineral. Due to the abundance of

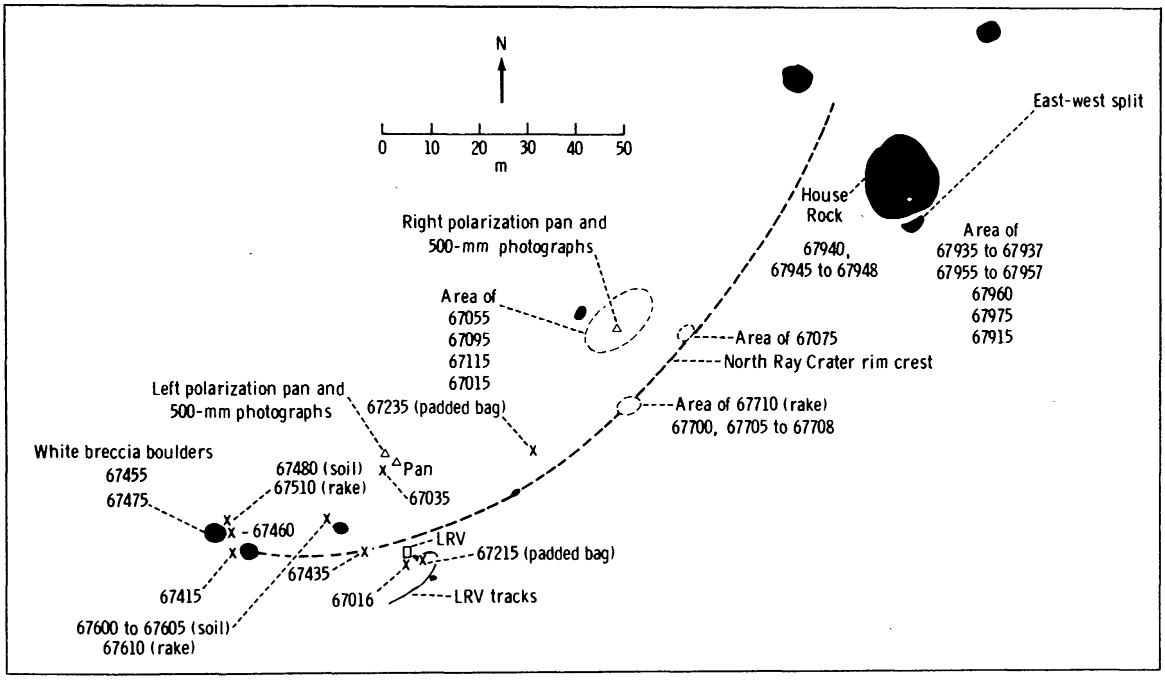

Fig. 2. Map of Station 11 at the Apollo 16 landing site. The dashed line denotes the rim crest of North Ray Crater. From AFGIT (1972).

spinel troctolites in Apollo 16 and Luna 20 soils, those authors indicate that such materials are a widespread lithology in the lunar highlands. Due to the low-Fe/(Fe + Mg) ratio, they suggest that the spinel troctolites are among the most primitive materials returned from the moon. Electron microprobe analyses of small samples from these inhomogeneous rocks yielded K concentrations which differ considerably from the accurate, whole-rock values shown in Table 1 for 60618 and 65785 (Dowty *et al.*, 1974). Comparison of Th and U concentrations of the spinel troctolite (65785) with previously studied Apollo 16 materials indicates a similarity with 62295, a "very high alumina" rock that has been called a troctolite and used as an important constituent of a mixing model study of lunar highland rock types (Schonfeld, 1974). Due to the similarity of composition and the primitive nature of the materials, anorthosites 65785 and 62295 will probably be important samples for future investigations. Additionally, we note that 60618 is unique in our suite of Apollo 16 samples in that it exhibits the lowest Th/U ratio (2.3) that we observed in all Apollo 16 materials, although the K/U ratio is similar to that of other Apollo 16 samples.

Primordial radioelements—Apollo 17

Special samples were collected during EVA-2 and EVA-3 at Taurus-Littrow by the use of a long-handled sampler (LRV sampler). Such samples were necessarily "grab" samples due to the restricted view of the sample area by the crew, who did not dismount from the LRV. Two pairs of LRV samples are included in this study and are indicated by underlines on the traverse map shown

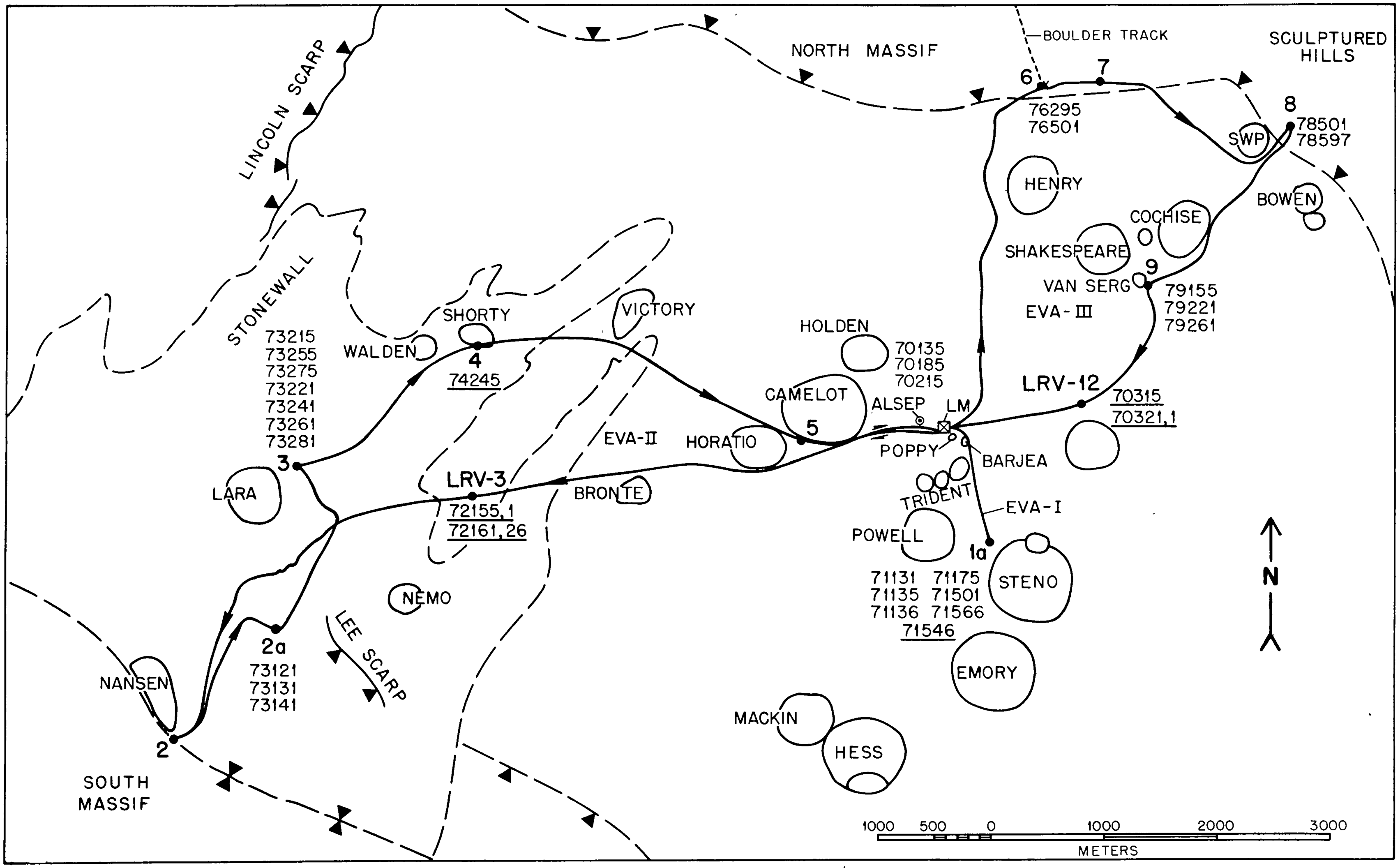

Fig. 3. Traverse map of the Taurus-Littrow region. Bold-faced numerals indicate sampling stations. Underlined numbers denote samples analyzed in this study; data on the other samples listed were reported by Eldridge *et al.* (1974).

in Fig. 3. Each pair consisted of a soil and a rock and were collected at LRV-3 during EVA-2 and at LRV-12 during EVA-3.

The LRV-3 sample stop was at a site in the dark mantle between the main body of light mantle and a finger of light mantle that lies to the southeast. The area resembles the dark-mantle surface at LRV-1 between Horatio and Bronte craters, rather than the dark-mantle surface in the vicinity of the lunar module (LM) or Station 1a. The rock sample at LRV-3 (72155,1) is a medium-grained basalt from a cluster of rocks on the surface that is not clearly related to any crater. The soil sample (72161,26) is typical of the surface material in a dark-mantle area free of large blocks (ALGIT, 1973). The LRV-12 sample stop is located about $\frac{1}{3}$ of a crater diameter out from the rim of Sherlock Crater in an area mapped as dark mantle. Due to the proximity to Sherlock Crater, it was speculated (ALGIT, 1973) that the LRV-12 rock sample (70315) is a subfloor basalt excavated from a depth of 50–90 m by the Sherlock event. The accompanying soil (70321,1) is probably representative of the dark-mantle surface (ALGIT, 1973).

The other two Apollo 17 samples in this study are: 71546, a fine-grained basalt fragment from the rake sampling about 150 m from the northwest rim of Steno Crater at Station 1a; and 74245, a fine-grained basalt from the end of the Station 4 trench in the south rim crest of Shorty Crater. Field geological studies indicate the wall, rim, and flank materials are Shorty Crater ejecta derived largely from materials above the subfloor, with basalt blocks at Station 4 coming from the subfloor materials.

Results of the radioelement concentrations in our current Apollo 17 samples are presented in Table 2 along with average values for the fine-, medium-, and coarse-grained basalts from our previous study (Eldridge *et al.*, 1974). The basalts in this group all have characteristically low values of Th/U that average 2.8, whereas the K/U ratio varies with texture. The previous K/U average for fine-grained basalts was 2930, 4015 for medium-grained, and 4590 for coarse-grained basalt. Note the K/U ratio for 74245 in Table 2 of 4770. Such a high value

Table 2. Apollo 17 radioelement concentrations.

Sample number	Type	K(ppm)	Th(ppm)	U(ppm)	Th/U	K/U
6-sple. avg.*	F. basalt	382	0.40	0.14	3.00 ± .26	2930 ± 260
74245,0	F. basalt	620 ± 30	0.40 ± .03	0.13 ± .02	3.08 ± .53	4770 ± 770
71546,0	F. basalt	500 ± 25	0.40 ± .03	0.15 ± .02	2.67 ± .41	3333 ± 475
4-sple. avg.*	M. basalt	444	0.38	0.11	3.48 ± .24	4015 ± 240
72155,1	M. basalt	525 ± 25	0.36 ± .02	0.13 ± .01	2.77 ± .26	4040 ± 365
6-sple. avg.*	C. basalt	495	0.34	0.11	3.12 ± .16	4590 ± 250
70315,0	C. basalt	400 ± 20	0.27 ± .02	0.10 ± .01	2.70 ± .34	4000 ± 450
70321,1	< 1-mm fines	595 ± 30	0.73 ± .07	0.26 ± .03	2.81 ± .42	2290 ± 290
72161,26	< 1-mm fines	795 ± 40	1.47 ± .08	0.45 ± .04	3.27 ± .34	1770 ± 180

*Eldridge *et al.* (1974).

would classify this Shorty Crater basalt into the same deep subfloor basalt flow as most of the Apollo 17 coarse-grained basalts. Sample 71546, with its fine-grained texture and K/U value of 3333, fits the previous classification for the uppermost basalt unit. Sample 70315, the LRV-12 rock that probably was ejected (ALGIT, 1973) from a depth of 50–90 m, has a K/U value that would tend to classify it as an intermediate basalt flow sample, even though its texture would place it in a deeper zone.

The pattern of primordial radioelement concentrations in the two soils of this study fits the field geology interpretation in the case of the LRV-12 soil 70321. The K, Th, and U concentrations are very similar to other "dark-mantle soils" such as 71131 and 71501 that we previously studied. However, soil 72161 collected at LRV-3 has a pattern that more closely resembles that of North Massif soils 76501 and 78501 from our earlier studies (Eldridge *et al.*, 1974) than it does those of the dark-mantle materials. This finding is in good agreement with the results of Rhodes, *et al.* (1974) who categorized Apollo 17 soils into three distinct chemical groups and classified sample 72161,6 as a "North Massif" type of soil.

Potassium—uranium systematics

Early studies of radioactivity in lunar samples indicated a distinct difference in the ratio of K to U in comparison with chondrites and terrestrial rocks (O'Kelley *et al.*, 1970a,b, 1971). In those mare basin samples, the K/U ratio fell within a narrow range of 1300–3200, even though the potassium concentration varied over the range of ~500–20,000 ppm. Figure 4 indicates the mass ratio K/U as a function of potassium concentration for all six Apollo sampling missions in relation to similar values for terrestrial and meteoritic materials. In contrast to the narrow K/U range for earlier missions, basalts from the Apollo 15 and 17 missions form a unique clustering that overlaps the eucrite zone.

It is also apparent that a trend line exists between the coordinates of the Apollo 17 basalts on the one hand and the point labeled KREEP on the other. Note that Apollo 17 soils and breccia fall on such a trend line at points intermediate to the end-members. Schonfeld (1974) investigated the possibility of highland rocks such as "very high alumina basalts" or "low-potassium Fra Mauro basalts" being mixtures by a mixing model calculation using up to 27 chemical elements. He showed that it is *possible* to generate such rock types by mixing dunite, "anorthosite," and KREEP. He developed a trend line with KREEP and (spinel) troctolite as end-members of the mixtures. It is interesting to note that the two important Apollo 17 rake samples—60618, with K/U = 2390 and K = 670 ppm; and 65785, with K/U = 1910 and K = 1850 ppm—fall on a trend line intersecting the coarse-grained Apollo 17 basalts and KREEP. In addition to the variations among basalts, soils and breccias for the Apollo 17 samples discussed above, the textural variations in the Apollo 17 basalts are reflected in the near-vertical clustering of K/U values at the potassium concentration of ~300–600 ppm. The range of K/U for such basalts is 1680–5000.

The later Apollo missions have provided a unique suite of samples that confirm

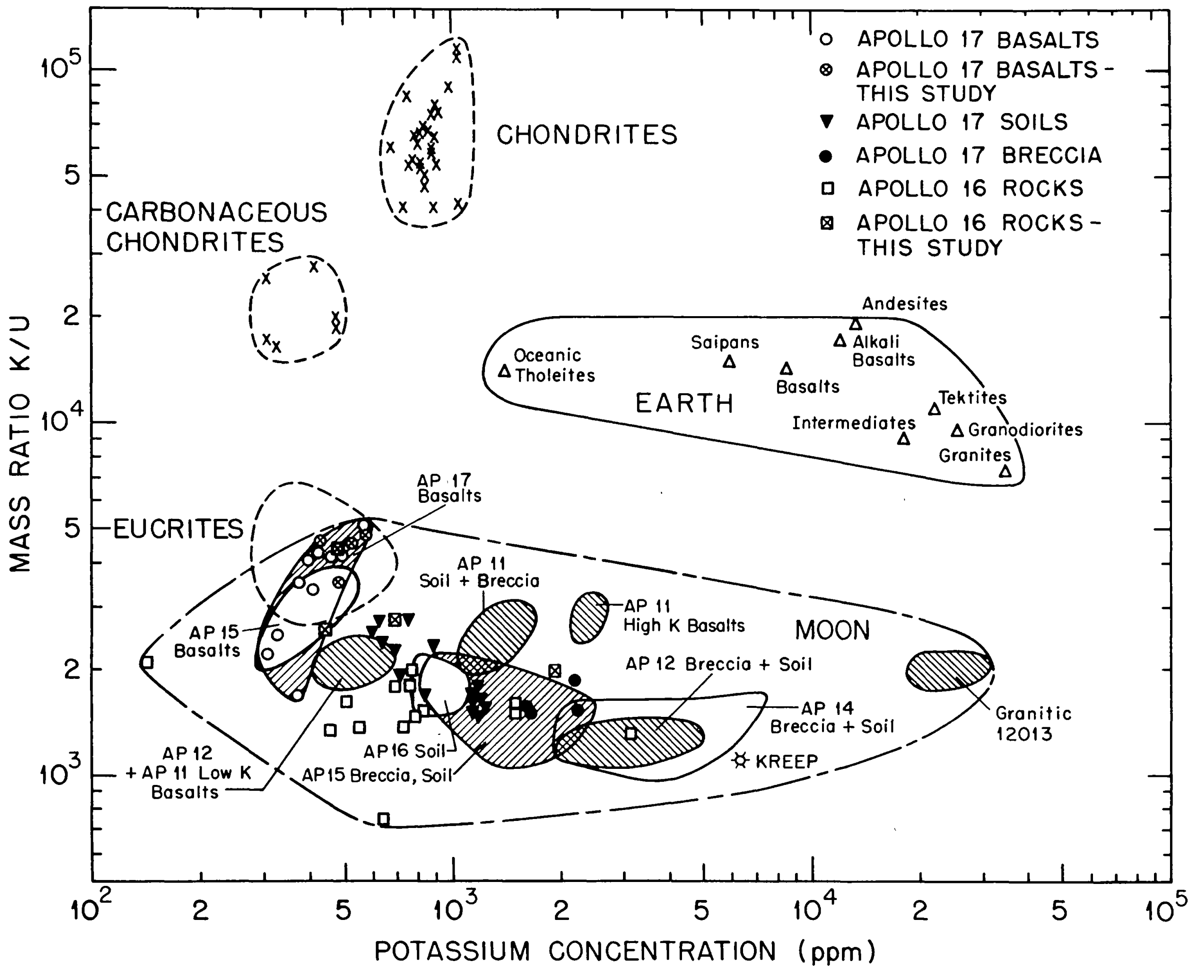

Fig. 4. Values of the mass ratio K/U as a function of the concentration of K. Apollo 16 and 17 materials appear to follow a trend line with basalts and KREEP as endmembers.

the extreme differentiation in lunar crustal materials. The primordial radioelement relationships may be coupled with past and future orbital γ-ray and X-ray experiments to delineate the crustal distributions of these important materials.

Cosmogenic radionuclides

The $2\frac{1}{2}$ yr between collection of samples and the measurements reported here precluded a determination of short-lived radionuclides. However, 312-day ^{54}Mn was detected in many of the Apollo 16 samples and all of the Apollo 17 samples, in addition to 2.6-yr ^{22}Na and 7.4×10^5-yr ^{26}Al. Results on these three cosmogenic species are summarized in Table 3.

As mentioned earlier, Descartes soils 63320 and 63340 were collected inside a hole at the south end of Shadow Rock, in an attempt to obtain permanently shadowed soil. Field geology studies (ALGIT, 1972) suggested that the samples may not have been completely shielded. Additionally, Imamura *et al.* (1974) found

Table 3. Concentrations (dpm/kg) of cosmogenic radionuclides.*

Sample number	Mass(g)	^{22}Na	^{26}Al	^{54}Mn
Descartes				
60618,0	20.5	45 ± 10	170 ± 9	—
63320,17	50.0	25 ± 5	69 ± 3	40 ± 20
63340,18	50.0	28 ± 6	91 ± 4	$\leqslant 50$
65785,0	5.0	45 ± 35	59 ± 6	—
67016,2	481.7	35 ± 3	88 ± 4	15 ± 10
67115,9	161.6	39 ± 4	67 ± 3	30 ± 15
Taurus-Littrow				
70315,0	147.4	82 ± 8	67 ± 4	165 ± 10
70321,1	99.8	130 ± 10	114 ± 8	195 ± 20
71546,0	148.7	94 ± 7	70 ± 3	165 ± 30
72155,1	160.6	68 ± 5	54 ± 3	125 ± 10
72161,26	99.7	190 ± 12	166 ± 10	220 ± 20
74245,0	61.4	28 ± 2	30 ± 2	60 ± 12

*Concentrations of ^{22}Na and ^{54}Mn corrected for decay to 23 April, 1972 and 14 December, 1972 for Apollo 16 and 17 samples, respectively.

that the concentration of 3.7×10^6-yr ^{53}Mn in sieved soil 63321 was higher than the production expected from galactic cosmic rays alone.

The short half-life of ^{22}Na (2.6 yr) makes it a useful indicator of recent solar cosmic-ray exposure, while ^{26}Al (7.4×10^5 yr) measures exposure history over a longer time scale. An appropriate comparison standard was the ^{22}Na concentration of 63501 determined previously (Eldridge *et al.*, 1973a). Surface soil 63501 was taken about 8 m from Shadow Rock and is assumed to have experienced negligible shielding from the sun (AFGIT, 1972). After correction for galactic cosmic-ray production of ^{22}Na in 63501 and 63320 we estimated that 63320 was exposed to about 20% of the solar cosmic-ray flux seen by exposed soil 63501, i.e. the shadowed soil 63320 was about 80% shielded from the sun.

The saturated activity of ^{26}Al in 63320 was taken to be 220 dpm/kg, the concentration found for 63501. After correction for galactic cosmic ray production and for the 20% solar cosmic ray exposure, an excess of about 20 dpm/kg of ^{26}Al remained over that expected for 63320 in its present location. This excess may be attributed to decay of initially saturated ^{26}Al over a period of about 3 m.y. These results are consistent with the conclusion that the shielding geometry of Shadow Rock was established about 3 m.y. ago, and afterward the shielding of 63320 with respect to solar cosmic rays was only about 80%. Such a conclusion is in qualitative agreement with the ^{53}Mn data of Imamura *et al.* (1974). The time scale is difficult to establish with precision, because the ^{26}Al concentrations of Table 3 show that the top layer of shadowed soil (63320) and the reference soil below (63340) were slightly mixed during sampling operations.

Earlier studies of Apollo 16 samples by Clark and Keith (1973) and by Eldridge *et al.* (1973a) showed that several rocks in the vicinity of North Ray Crater were unsaturated in ^{26}Al content. These results were confirmed by Yokoyama *et al.* (1974). We have applied the criteria for saturation of ^{26}Al suggested by Keith and Clark (1974) and by Yokoyama *et al.* (1974) to the Apollo 16 rocks of Table 3. Rock 67115,9 was found to be unsaturated in ^{26}Al, confirming the earlier measurements by Clark and Keith (1973) on the same sample. Although no chemical analysis data exist for light-matrix breccia 67016,2 it also appears to be unsaturated in ^{26}Al.

Two small anorthositic rocks from the Apollo 16 rake collection were studied. Sample 60618, 20.5 g, appears to be saturated in ^{26}Al, while 67785, 5.0 g, is considerably undersaturated. However, the small sample sizes and the preliminary nature of the chemical analysis data introduce some uncertainty into these conclusions.

The Apollo 17 collection included two pairs of samples from LRV stations (cf. Fig. 3). Coarse basalt 70315 and reference soil 70321 were taken at Station LRV-12, and medium basalt 72155 and soil 72161 from LRV-3. Both of these soils were poorly documented, and it is only known that they were taken from the top few centimeters of the surface (ALGIT, 1973). Cosmogenic radionuclide concentrations in these samples closely resemble those of other Apollo 17 bulk surface soils discussed in detail by O'Kelley *et al.* (1974). The effects of the August, 1972 solar flare are apparent to some extent in all Apollo 17 samples reported in Table 3. The ^{26}Al concentration in basalts 70315 and 72155 is below the expected saturation value, indicating a short exposure age or shielding due to soil cover. The ^{22}Na data are difficult to evaluate but are consistent with partial shielding.

Rock 71546 is a fine-grained basalt, one of the larger fragments collected as part of the rake sample at Station 1a. Most of the rake fragments were partially or even completely buried prior to collection. Sample 71546 shows evidence of partial shielding from the intense solar event of August, 1972, and so was probably one of the covered, near-surface samples.

Sample 74245 was a small fragment of fine-grained basalt from Station 4. It was collected with a sample of gray soil from the ends of the trench crossing the zone of "orange" soil. No documentation of this rock is available, but its cosmogenic radionuclide content suggests that 74245 was buried several centimeters deep before it was removed from the sampling trench.

Acknowledgments—The authors gratefully acknowledge the assistance and advice provided by the Lunar Sample Analysis Planning Team. We wish to thank the staff of the Curator's Office, NASA Johnson Space Center, especially J. O. Annexstad and M. A. Reynolds, for their aid in rapidly and efficiently processing the samples for this study.

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