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NUCLEAR CHEMISTRY OF RETURNED LUNAR SAMPLES:
NUCLIDE ANALYSIS BY GAMMA-RAY SPECTROMETRY

NASA Order T-2400A
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for the
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The first part of the progress report for this period is a brief account of measurements on an important sample, not yet prepared for publication. The second part consists of two reprints and one preprint of completed work, published or in press.

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KREEP is ubiquitous at the Apollo 15 site and has been estimated\textsuperscript{2,3} to be present at concentrations of more than 20% in some Apollo 15 soils and breccias. Basalt fragments with fresh subophitic textures and characteristic KREEP chemistry were discovered in soil 15023 by Meyer\textsuperscript{4}. Thin sections of rake samples 15382 and 15386 have textures expected for KREEP basalts, and we have now completed measurements on the radionuclide contents of both.

Earlier we reported\textsuperscript{5} our results on 15382, and in the present reporting period we completed measurements on 15386. In Table 1 we compare K, Th, U, and $^{26}$Al concentrations of 15382, 15386, 14310, and KREEP. Our data on 15382 and 15386 were determined on very small samples with our nondestructive gamma-ray spectrometry method.

The primordial radioelement concentrations of 15382 and 15386 are similar, and they, in turn, closely resemble the concentration pattern of KREEP. No other lunar rock type has K, Th, and U concentrations similar to KREEP. Thus, both 15382 and 15386 could be pure KREEP or a mixture with only slight contamination. It is difficult to be more quantitative in comparing the data of Table 1, because the variability of primordial radioelement concentrations in KREEP is about 30%.

The information presented here shows that 15382 and 15386 are important samples for additional studies of chemistry, mineralogy, and radiogenic age.

REFERENCES AND NOTES

1. NASA Johnson Space Center.


Table 1. Radionuclide Concentrations

<table>
<thead>
<tr>
<th>Sample</th>
<th>weight (g)</th>
<th>K (ppm)</th>
<th>U (ppm)</th>
<th>Th (ppm)</th>
<th>$^{25}_{Al}$ (dpm/kg)</th>
<th>K/U</th>
<th>Th/U</th>
</tr>
</thead>
<tbody>
<tr>
<td>15386</td>
<td>6.41</td>
<td>4970±200</td>
<td>3.30±0.15</td>
<td>11.8±0.3</td>
<td>94±9</td>
<td>1500</td>
<td>3.6</td>
</tr>
<tr>
<td>15382$^a$</td>
<td>2.24</td>
<td>4900±500</td>
<td>3.1±0.4</td>
<td>10.5±0.5</td>
<td>74±20</td>
<td>1610</td>
<td>3.4</td>
</tr>
<tr>
<td>14310$^b$</td>
<td>--</td>
<td>4250</td>
<td>3.06</td>
<td>10.8</td>
<td>--</td>
<td>1390</td>
<td>3.5</td>
</tr>
<tr>
<td>KREEP$^c$</td>
<td>--</td>
<td>5200</td>
<td>4.5</td>
<td>17.2</td>
<td>--</td>
<td>1160</td>
<td>3.8</td>
</tr>
</tbody>
</table>

$^a$Schonfeld, et al., ref. (5).

$^b$Average of values from the literature.

$^c$Schonfeld and Meyer, ref. (3).
II. REPRINTS AND PREPRINT OF COMPLETED WORK

This section of the report contains the following articles, published or in press.


Primordial radioelement concentrations in rocks and soils from Taurus-Littrow*

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Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

Abstract—Primordial radioelement (K, Th, and U) concentrations were determined nondestructively by gamma-ray spectrometry in thirteen soil and thirteen rock samples from the returned sample collection from Taurus-Littrow. Soil samples investigated were: 71131, 71501, 73121, 73131, 73141, 73221, 73241, 73261, 76501, 78501, 79221, and 79261. Rock samples were: 70135, 70185, 70215, 4, 71135, 71136, 71175, 71566, 73215, 73255, 73275, 76295, 78597, and 79155.

Concentrations of K, Th, and U in the soil samples were found to have a narrow range of values for seven light-mantle soils that were distinctly different from those values for the other six soils in this study. The Th content of the light soils is the same as that found from the orbiting gamma-ray spectrometer determination of Trombka et al. (1973). Little or no variation of primordial radioelement content was observed as a function of depth, color, or texture in trench samples.

Variations of Th/U and K/U were observed in three types of basaltic rocks. These observations lend geochemical evidence to the speculation that two or three separate subfloor basalt units were sampled at the Taurus-Littrow site. Correlation of K/U mass ratios with potassium content of the Apollo 17 materials indicate a possible two component mixing of subfloor basalt and KREEP as end members to produce soil and breccias at Taurus-Littrow.

INTRODUCTION

The returned sample collection from Taurus-Littrow contained materials from three of five expected geologic units. Soil and rock samples were obtained that characterize the subfloor unit and its associated regolith, the light mantling material, and the highlands material from the North and South Massifs. Nondestructive gamma-ray spectrometry was used on a suite of rock and soil samples from each of the geologic units at Taurus-Littrow to determine the content of primordial radioelements (K, Th, and U) along with radionuclides produced by galactic and solar proton activation. Results and interrelationships of the activation product content of the same group of samples reported here are presented in a companion report in this volume (O'Kelley et al., 1974).

A measure of the total radioactivity of the moon can yield important information concerning its thermal history. Schonfeld (1974a) estimated that the moon's upper mantle (or that part of the moon where most lunar samples were derived) has an average U concentration of 0.085 ppm. Wänke et al. (1973, 1974) calculated an average U concentration of 0.086 and 0.077 ppm for the part of the moon that underwent magmatic differentiation. Both Schonfeld (1974a) and Wänke et al. (1973) indicated that their calculated U average concentrations were consistent

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with experimentally determined heat flow values from the Apollo 15 and 17 missions. In addition, the Th values reported here served as an important correlation with orbiting gamma-ray spectrometer measurements (Trombka et al., 1973).

Part of the results from this study were obtained during the preliminary examination period of the Apollo 17 mission and are included in the LSPET (1973a) report. The present report includes additional results and further characterization of Th—U and K—U correlations that had been observed in samples from other Apollo landing sites.

**EXPERIMENTAL METHODS**

The gamma-ray spectrometer system used in these studies has been previously described (O’Kelley et al., 1970) and (Eldridge et al., 1973). Resolution of the complex gamma-ray spectra from both the coincidence matrix and from the summed output of the single detectors was accomplished with ALPHA-M, a program for quantitative radionuclide determination (Schonfeld, 1967). Accurate replicas of all rock samples in this study were made from rock models, provided by the Curator’s Office, Johnson Space Center. Standard gamma-ray spectrum libraries were acquired by measurement of accurate replicas (containing standards for each nuclide sought) under the same experimental conditions as the lunar samples. Error values listed for all determinations in this series are overall estimates including counting statistics and system calibrations. Sample weights ranged from 24 to 3600 g for the rock samples with the median weight near 400 g. Soil samples were nominally 50 or 100 g aliquots of <1 mm fines. Density determinations for rock samples were made by determining the volume of thin-shelled aluminum replicas by pressing aluminum foil around the rock models to make hollow shells. From the weight of the lunar sample and the volume of its replica shell, density values were calculated.

**RESULTS AND DISCUSSION**

Rock and soil samples from a number of sampling stations were studied in order to characterize the primordial radioelement content of each of the geologic units at Taurus-Littrow. Figure 1 shows a diagram of the Apollo 17 landing site with the traverses, sampling stations, and major physiographic features indicated. Sampling stations are indicated by the bold-face Arabic numerals. Samples measured in this study are listed near the corresponding sampling station. Primordial radioelement concentrations for the 26 samples in this study are shown in Table 1.

The Apollo Field Geology Investigation Team speculated that sampled subfloor basalts were derived from 20 to 130 m depths, and that the stratigraphically lowest basalt unit was vesicular and coarse-grained. Other stratigraphic units graded upward to coarse-grained porphyritic, vesicular fine-grained, and finally aphanitic basalts in the shallowest recognizable type (AFGIT, 1973). Table 2 contains average primordial radioelement concentrations for 16 basalt samples, from this study and LSPET (1973a), grouped according to fine-, medium-, and coarse-grained classifications of LSPET (1973a). The average value for the K/U of fine-grained basalts is significantly different from that ratio in the medium and coarse basalts. This geochemical difference observed here in the discrepancy of
Fig. 1. Apollo 17 landing area in the Taurus-Littrow region. Bold-faced Arabic numerals indicate sampling stations and five digit numerals indicate sample numbers with soils, having a 1 as the final digit. The coordinates for the lunar module (LM) are: latitude 20°10'N and longitude 30°46'E.
Table 1. Primordial radioclenlent concentrations in Apollo 17 samples.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Density (g/cm³)</th>
<th>Type*</th>
<th>K (ppm)</th>
<th>Th (ppm)</th>
<th>U (ppm)</th>
<th>Th/U</th>
<th>K/U</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rocks</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>70135</td>
<td>3.0</td>
<td>CB</td>
<td>500 ± 30</td>
<td>0.31 ± .02</td>
<td>0.12 ± .01</td>
<td>2.6</td>
<td>4170</td>
</tr>
<tr>
<td>70185</td>
<td>3.0</td>
<td>CB</td>
<td>420 ± 35</td>
<td>0.30 ± .03</td>
<td>0.10 ± .02</td>
<td>3.0</td>
<td>4200</td>
</tr>
<tr>
<td>70215</td>
<td>3.3</td>
<td>FB</td>
<td>220 ± 64</td>
<td>0.36 ± .03</td>
<td>0.13 ± .03</td>
<td>2.8</td>
<td>2500</td>
</tr>
<tr>
<td>71135</td>
<td>1.9</td>
<td>FB</td>
<td>350 ± 40</td>
<td>0.60 ± .05</td>
<td>0.14 ± .03</td>
<td>4.3</td>
<td>2214</td>
</tr>
<tr>
<td>71176</td>
<td>2.4</td>
<td>FB</td>
<td>370 ± 100</td>
<td>0.46 ± .06</td>
<td>0.22 ± .05</td>
<td>2.1</td>
<td>1680</td>
</tr>
<tr>
<td>71175</td>
<td>2.4</td>
<td>MB</td>
<td>560 ± 28</td>
<td>0.39 ± .02</td>
<td>0.11 ± .01</td>
<td>3.5</td>
<td>5060</td>
</tr>
<tr>
<td>71566</td>
<td></td>
<td>CB</td>
<td>450 ± 20</td>
<td>0.31 ± .01</td>
<td>0.09 ± .008</td>
<td>3.4</td>
<td>4890</td>
</tr>
<tr>
<td>73215</td>
<td>2.5</td>
<td>BR</td>
<td>1665 ± 85</td>
<td>4.05 ± .20</td>
<td>1.10 ± .05</td>
<td>3.7</td>
<td>1514</td>
</tr>
<tr>
<td>73255</td>
<td>2.4</td>
<td>BR</td>
<td>1590 ± 80</td>
<td>3.47 ± .17</td>
<td>1.00 ± .05</td>
<td>3.5</td>
<td>1590</td>
</tr>
<tr>
<td>73275</td>
<td>2.2</td>
<td>BR</td>
<td>2240 ± 112</td>
<td>4.53 ± .23</td>
<td>1.20 ± .06</td>
<td>3.8</td>
<td>1867</td>
</tr>
<tr>
<td>76295</td>
<td>2.4</td>
<td>BR</td>
<td>2270 ± 114</td>
<td>5.20 ± .27</td>
<td>1.50 ± .08</td>
<td>3.5</td>
<td>1510</td>
</tr>
<tr>
<td>78579</td>
<td>2.6</td>
<td>FB</td>
<td>380 ± 20</td>
<td>0.38 ± .02</td>
<td>0.11 ± .01</td>
<td>3.4</td>
<td>2454</td>
</tr>
<tr>
<td>78655</td>
<td>2.6</td>
<td>CB</td>
<td>440 ± 30</td>
<td>0.32 ± .02</td>
<td>0.11 ± .01</td>
<td>2.9</td>
<td>4000</td>
</tr>
<tr>
<td>Soils (&lt;1 mm fines)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>71131</td>
<td>SS</td>
<td>625 ± 30</td>
<td>0.67 ± .05</td>
<td>0.23 ± .02</td>
<td>2.9</td>
<td>2720</td>
<td></td>
</tr>
<tr>
<td>71501</td>
<td>RS</td>
<td>585 ± 30</td>
<td>0.75 ± .04</td>
<td>0.23 ± .02</td>
<td>3.3</td>
<td>2540</td>
<td></td>
</tr>
<tr>
<td>11121</td>
<td>SS</td>
<td>1160 ± 60</td>
<td>2.63 ± .13</td>
<td>0.72 ± .04</td>
<td>3.7</td>
<td>1610</td>
<td></td>
</tr>
<tr>
<td>71311</td>
<td>SS</td>
<td>1160 ± 60</td>
<td>2.24 ± .11</td>
<td>0.63 ± .03</td>
<td>3.6</td>
<td>1840</td>
<td></td>
</tr>
<tr>
<td>71411</td>
<td>TB</td>
<td>1130 ± 60</td>
<td>2.25 ± .11</td>
<td>0.63 ± .03</td>
<td>3.6</td>
<td>1790</td>
<td></td>
</tr>
<tr>
<td>73221</td>
<td>TT</td>
<td>1180 ± 60</td>
<td>2.13 ± .11</td>
<td>0.63 ± .03</td>
<td>3.4</td>
<td>1870</td>
<td></td>
</tr>
<tr>
<td>73241</td>
<td>TM</td>
<td>1220 ± 60</td>
<td>2.25 ± .11</td>
<td>0.64 ± .03</td>
<td>3.5</td>
<td>1910</td>
<td></td>
</tr>
<tr>
<td>73261</td>
<td>TB</td>
<td>1050 ± 60</td>
<td>2.40 ± .12</td>
<td>0.67 ± .04</td>
<td>3.6</td>
<td>1630</td>
<td></td>
</tr>
<tr>
<td>73281</td>
<td>TB</td>
<td>1180 ± 120</td>
<td>2.33 ± .11</td>
<td>0.58 ± .04</td>
<td>4.0</td>
<td>2030</td>
<td></td>
</tr>
<tr>
<td>75501</td>
<td>RS</td>
<td>930 ± 50</td>
<td>1.39 ± .14</td>
<td>0.58 ± .04</td>
<td>3.6</td>
<td>2370</td>
<td></td>
</tr>
<tr>
<td>75851</td>
<td>RS</td>
<td>770 ± 40</td>
<td>1.11 ± .11</td>
<td>0.28 ± .03</td>
<td>4.0</td>
<td>2750</td>
<td></td>
</tr>
<tr>
<td>79221</td>
<td>TT</td>
<td>700 ± 40</td>
<td>1.12 ± .06</td>
<td>0.26 ± .03</td>
<td>3.1</td>
<td>1940</td>
<td></td>
</tr>
<tr>
<td>79261</td>
<td>TB</td>
<td>700 ± 40</td>
<td>1.08 ± .05</td>
<td>0.31 ± .02</td>
<td>3.5</td>
<td>2260</td>
<td></td>
</tr>
</tbody>
</table>

*CB = coarse basalt, FB = fine basalt, MB = medium basalt, BR = breccia, TT = trench top, TM = trench middle, TB = trench bottom, RS = rake soil, SS = surface soil.

K/U grading from coarse-grained to the fine-grained basalts would tend to verify the speculation of separate flow units in the mare basalts from the subfloor at Taurus-Littrow.

Basalt samples have Th/U ratios distinctly lower than the usual lunar and terrestrial value of ~3.8. Medium and coarse basalt have K/U ratios of ~4300; differing greatly from the previously observed lunar average of ~2500 (Schonfeld, 1974a). These geochemical differences indicate not only that the basalt flows filling the Taurus-Littrow Valley are different in themselves, but also different from previously sampled basalts from other Apollo sites. Duncan et al. (1974a) have noticed differences in K/Zr ratios of Apollo 17 samples compared to a near constant ratio they observed in Apollo 12, 14, and 15 samples. They correlated
Primordial radioelement concentrations in rocks and soils from Taurus-Littrow

Table 2. Average primordial radioelement content of Apollo 17 rocks.

<table>
<thead>
<tr>
<th>Rock type</th>
<th>No.</th>
<th>K (ppm)</th>
<th>Th (ppm)</th>
<th>U (ppm)</th>
<th>Th/U</th>
<th>K/U</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fine basalt</td>
<td>6</td>
<td>382</td>
<td>0.40</td>
<td>0.14</td>
<td>3.00 ± 0.24</td>
<td>2930 ± 260</td>
</tr>
<tr>
<td>Medium basalt</td>
<td>4</td>
<td>444</td>
<td>0.38</td>
<td>0.11</td>
<td>3.48 ± 0.24</td>
<td>4015 ± 240</td>
</tr>
<tr>
<td>Coarse basalt</td>
<td>6</td>
<td>495</td>
<td>0.34</td>
<td>0.11</td>
<td>3.12 ± 0.16</td>
<td>4590 ± 252</td>
</tr>
<tr>
<td>Breccia</td>
<td>4</td>
<td>1940</td>
<td>4.34</td>
<td>1.20</td>
<td>3.62 ± 0.12</td>
<td>1620 ± 57</td>
</tr>
</tbody>
</table>

K/Zr ratios with TiO₂ contents and invoked a two-stage origin for high-titanium mare basalts.

Primordial radioelement concentrations in the four breccia samples of this study are approximately an order of magnitude greater than any of the three basalt types. In addition, the Th/U and K/U values are near normal and overlap those from Apollo 15 and 16 breccia samples (see Fig. 2 and Clark and Keith, 1973). Since the primordial radioelement content of the breccias is approximately twice that of even the light soils it appears that the breccias could not have been formed from simple impact induration of the upper regolith.

Fig. 2. Values of the mass ratio K/U as a function of the concentration of K. Apollo 17 basalts, soils, and breccias appear to fall on a line that intersects KREEP.
The U average value of 0.11 ppm shown in Table 2 for basalts is not greatly different from the predicted upper mantle average of 0.085 ppm as given by Schonfeld (1974a), and from the whole moon (or the part that underwent magmatic differentiation) average of 0.077 ppm calculated by Wänke et al. (1974).

Figure 2 shows a comparison of the mass ratio K/U and potassium concentration of Apollo 17 samples with earth, meteorite, and other lunar samples. It can be seen that the "moon" values fall in a region distinctly different from "earth" and most meteorite ratios. Apollo 17 basalts occupy a region which overlaps that of Apollo 15 basalts.

Grouping of the Apollo 17 basalts, soils, and breccias in the K—U systematic diagram of Fig. 2 indicates a possible two component mixing model for the soils and breccias; with coarse basalts and KREEP as the end members. In spite of the limited number of samples, this observation would lead to the speculation that breccias and soils at the Apollo 17 site may be indurated products of KREEP and basaltic materials with the KREEP component less abundant than that of the Fra Mauro breccias. This speculation is a very limited interpretation and is not as complete as the more extensive Apollo 17 regolith studies by several other groups (Duncan et al., 1974b; Rhodes et al., 1974; Schonfeld, 1974b).

The radioclement content of soil samples from the light-mantle unit differs greatly from that of soils collected from other points in the valley floor. Seven light-mantle soils in this study yielded average concentrations (ppm) of 1.16, 2.32, and 0.64 for K, Th, and U, respectively. These concentrations are 2-3 times as great as those of the other six soils in the valley floor regolith samples from four different sampling stations.

The thorium concentration of 2.32 ppm for the light soil average compares favorably with the value of 2.2 ppm determined for a 5°x5° segment at Littrow by Trombka et al. (1973) in their orbital experiments. The orbital measurement covers a large area at a mare—highland interface; but Trombka et al. (1973) stipulate that the light soils are more typical of the general region covered by the orbital spectrometer. Since the orbital measurement averages the Th content of the entire 5°x5° segment, it is obvious that the dark valley soils cannot be a substantial fraction of the 5°x5° area because of the low Th content of the six dark- and intermediate-albedo soils.

Three light-mantle soils collected at Station 2a may be examined for near surface vertical variations. Samples 73121 and 73131 are surface soils at Station 2a, while 73141 was collected from the bottom of a 15 cm trench. There is no difference in potassium concentrations in the three soils and only slight elevation of Th and U in 73121 compared with 73131 and 73141. Field geology studies indicate that sample 73140 (the parent soil from which <1 mm fines 73141 were sifted) might be the most representative sample of light-mantle fines returned by the Apollo 17 mission (ALG1T, 1973). For this reason, analytical results for 73141 are especially useful as being representative of South Massif regolith.

Four samples from a trench in the light mantle at Station 3 were studied to determine any primordial radioclement variations with depth and with color variations within the 10 cm trench. It can be seen that there are no significant
differences in K, Th, or U concentrations in 73221, 73224, 73261, or 73281 even though these four samples cover several depth variations from the upper 0.5 cm of soil to 10 cm below the surface. The trench samples also include color variations from white to gray along with variable agglutinate content (LSPET, 1973a).

Another pair of trench samples shown in Table I are 79221 and 79261 from the top and bottom of a 17 cm trench exposed in the southeast flank of Van Serg Crater ejects blanket. The surface sample 79221 contains twice as much agglutinate (ubiquitous soil component consisting of mineral and lithic detritus bonded by grapelike clusters of brown glass) as the trench bottom, 79261, along with very different breccias (AFGIT, 1973). Again, this pair of samples shows no primordial radioelement differences in spite of major color, texture, and depth variations. This Station 9 trench pair is greatly different in chemical composition from the Stations 2n and 3 soils; and their K, Th, and U concentrations are similar to the dark soils at Taurus-Littrow.

A surface soil and a rake soil collected about 15 m apart at Station 1 (71131 and 71501) contain K, Th, and U concentrations that are similar to those of the basalts collected at the same location (71135, 71136, and 71175). In addition, these Station 1 soils are significantly lower in primordial radioelements than the other dark- or intermediate-albedo soils from Stations 6, 8, and 9 listed in Table I. The Th/U ratio for the Station 1 soils (2.91±0.33 and 3.26±0.33 for 71131 and 71501, respectively) is much lower than the ~3.8 value observed for previous Apollo soils. The low Th/U ratio together with the low overall primordial radioelement concentrations is a reflection of the high basaltic content of these soils. These dark soils from the valley floor contain the lowest primordial radioelement concentrations of all the other Apollo sampling sites (Eldridge et al. 1974).

Thorium to uranium ratios discussed in this paper are not as precise as those measured by some other analytical techniques, such as isotope dilution mass spectrometry. The accuracy is limited by uncertainties in both Th and U due to statistics of counting. However, the nondestructive nature of gamma-ray spectrometry measurements allows one to use samples large enough to average out sampling inhomogeneities. Nunes et al. (1974) reported U and Th concentrations of 0.313 and 1.134 ppm, respectively for soil sample 79221; whereas the corresponding values from Table I are 0.36±0.03 and 1.12±0.06 ppm. These comparisons are for the same size fraction (<1 mm fines) of soil 79221; whereas the corresponding values from Table I are 0.36±0.03 and 1.12±0.06 ppm. These comparisons are for the same size fraction (<1 mm fines) of soil 79221. Silver (1974) reported U and Th concentrations of 0.406 and 1.337 ppm, respectively for an unsieved soil, 78500.6. Uranium and thorium concentrations reported in Table I for 78501, the <1 mm size fraction from soil 78500, are 0.28±0.03 and 1.11±0.11 ppm, respectively. It is obvious that the differences are outside the reported errors.

Duncan et al. (1974b) studied both major and trace element concentrations in Apollo 17 regolith and noted considerable compositional variations related to selenographic position. In addition, those authors separated a dark-mantle soil, 75081, into four size fractions and noted a significant increase in a KREEP-rich component and an "orange soil" component in the finest fraction. The <300 mesh fraction of the <1 mm fines contained 40% more K, 54% more P, and 20% less Ti.
than the coarsest fraction of the <1 mm fines. In addition, Zn, Cu and Ni were present in a threefold increase over the coarse fractions.

Due to the compositional differences in a sieved sample in this work compared to the unsieved sample of Silver (1974) and to the significant correlation of a KREEP-rich component with the finest fraction of a dark-mantle soil by Duncan et al. (1974b), it can be postulated that analytical results for Apollo 17 regolith samples should be compared with caution. Large sample populations should be studied in order to average sampling defects. Silver (1974) reported average uranium and thorium levels in surface soils of the valley floor as 0.344 and 1.09 ppm, respectively. He reported that the average Th/U ratio of 3.14 was distinguishably lower than all Apollo mission sites except Apollo 16.

CONCLUSIONS

Primordial radionuclide determinations in this study have shown geochemical evidence in support of field geology speculation concerning layering in the subfloor basalt flows along with a possible correlation of magmatic fractionation of K/U as a function of depth. Elevated values of primordial radionuclide concentrations in breccia samples compared to the soils at Taurus-Littrow indicate that the breccias could not be formed from simple impact induration of the local regolith. On the other hand, similarities of the K, Th, and U content of the soils and basaltic rocks (as well as the similar Th/U ratio of ~3.1) indicate that the dark regolith in the Taurus-Littrow Valley may be the comminuted product of the underlying subfloor unit mixed with other components.

Correlation of a KREEP-rich component and an "orange soil" component with the finest fraction of <1 mm soil by Duncan et al. (1974b), coupled with an unresolved analytical uncertainty between an unsieved soil value of Silver (1974) and a <1 mm fraction of the same soil in this work, leads to the conclusion that Apollo 17 regolith samples are subject to sampling inhomogeneities. For this reason, care should be exercised in comparing analytical results of Apollo 17 regolith samples obtained with different size fractions.

Acknowledgments—The authors gratefully acknowledge the assistance provided by the Lunar Sample Analysis Planning Team in granting suitable sample allocations for this study. In addition, we wish to thank the Curator's staff at Johnson Space Center, especially J. O. Annexstad, R. S. Clark, and M. A. Reynolds, for the expeditious processing of the numerous samples in this work.

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Primordial radionuclide concentrations in rocks and soils from Taurus-Littrow


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Cosmogenic radionuclides in samples from Taurus-Littrow: Effects of the solar flare of August 1972*

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Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

Abstract—Cosmogenic radionuclide concentrations in a suite of Apollo 17 samples were determined nondestructively by use of gamma-ray spectrometers with high sensitivity and low background. Samples investigated were rocks 70135, 70185, 71135, 71175, 71566, 73215, 73255, 73275, 76295, 76597, and 79135; and <1 mm fines from soils 71131, 71501, 73121, 73131, 73141, 73221, 73241, 73261, 73281, 76501, 78501, 79221, and 79261. Cosmogenic radionuclides determined in this study were \(^{24}\)Na, \(^{26}\)Al, \(^{30}\)Si, \(^{34}\)V, \(^{53}\)Mn, \(^{57}\)Co, and \(^{10}\)Be.

The pattern of radionuclide concentrations observed in these Apollo 17 samples is quite distinct from that of any previous lunar sampling mission, due to proton bombardment by the intense solar flares of August 4–9, 1972. This intense proton irradiation made it possible for us to identify \(^{10}\)Be in a lunar sample for the first time.

Data on the \(^{57}\)Co and \(^{53}\)Mn contents of thin surface samples were used to calculate the proton flux and rigidity of the August, 1972 solar flare. We determined the integrated proton flux \(J_\gamma(\geq 10 \text{ MeV}) = 1.9 \times 10^{10} \text{ cm}^{-2}\) and for an energy spectrum expressed as the function \(E^{-\alpha}\), the shape parameter \(\alpha\) was found to be 1.92 ± 0.1. Thus, the solar cosmic ray event of August 1972 exhibited a much higher average proton energy than other flares of cycle 20, which had characteristic values of \(\alpha = 3.0\).

INTRODUCTION

The Apollo 17 voyage of exploration to the valley of Taurus-Littrow yielded the greatest scientific benefits of the six manned lunar landing missions. As a result of experience on previous Apollo flights, planning for Apollo 17 was characterized by a carefully selected landing site of great importance, a high degree of astronaut mobility, rapid scientific assessment both on the moon and on the earth, and a collection of well-documented samples representative of the geological complexities of the area. In addition, samples from the Taurus-Littrow Valley proved to be of great interest because, prior to collection, they had been subjected to proton bombardment by the intense sequence of solar flares of August 4–9, 1972. Data on radionuclide concentrations in thin surface samples offered the opportunity to determine the flux and energy spectrum of the solar flare protons, while samples from depth could be used to measure the influence of the galactic cosmic ray component.

As in our studies on samples from previous Apollo flights, we applied the methods of nondestructive gamma-ray spectrometry to the determination of radionuclides in samples from Taurus-Littrow. Measurements of cosmogenic radionuclides in 25 samples from Apollo 17 are reported below. Because several

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radionuclides have short half-lives, it was desirable to begin measurements as soon as possible. Our first samples were received six days after splashdown, and additional measurements were carried out during the preliminary examination. An early account of the results on some of these samples was given in LSPET (1973b), and a more complete account in O'Kelley et al. (1974).

Nondestructive gamma-ray spectrometry has proved to be valuable as an accurate and rapid method for scanning a large number of samples to determine concentrations of the primordial radioelements K, Th, and U in lunar samples. Data on the K, Th, and U contents of 26 samples from Taurus-Littrow and correlations with local terrain features are reported by Eldridge et al. (1974) in a companion paper of this volume.

**Experimental Procedures**

Two gamma-ray scintillation spectrometers were used for these analyses. The first was a Ge(Li) detector enclosed in a passive lead shield of 8 cm wall thickness. The detection efficiency at 1332 keV was 16% that of a 7.6×7.6 cm NaI(Tl) detector at a source-to-detector distance of 25 cm. Energy resolution expressed as full width at half-maximum counting rate at 1332 was 2.5 keV. Pulse-height distributions were recorded on a 4096-channel analyzer.

Most of the measurements were carried out with a scintillation gamma-ray spectrometer of low background, which contained two NaI(Tl) detectors, each 23 cm in diameter and 13 cm long, with a 10 cm pure NaI light guide. The detectors were operated in singles and coincidence modes with a large plastic scintillator in anticoincidence. Further background reduction was achieved with a massive lead shield 20 cm thick. The data acquisition system included a coincidence-anticoincidence logic circuit interfaced to a 4096-channel analyzer. Singles data from each NaI(Tl) detector were recorded, and coincidence events were sorted in a 63×63-channel matrix. The mechanical design and performance of this system have been discussed in detail by Eldridge et al. (1973).

Least-squares fitting of the gamma-ray spectra was performed on an IBM 360/91 computer with ALPHA-M, a program written by Schonfeld (1967). Standard gamma-ray spectrum libraries were acquired by measurement of replicas containing radionuclide standards under the same experimental conditions as for the lunar samples. Replicates fabrication, least-squares data analysis, and results on analyses of radionuclide test mixtures were discussed by O'Kelley et al. (1970) and by Eldridge et al. (1973). Errors listed in the tables below are overall estimates including counting statistics and uncertainties in the calibrations.

**Results and Discussion**

**Cosmogenic radionuclide concentrations**

Concentrations of cosmogenic radionuclides determined in this study are given in Table 1. Values for short-lived nuclides have been corrected for decay to 2300 GMT December 14, 1972, the approximate time of departure of the lunar module's ascent stage from the moon.

The general pattern of radionuclide concentrations in Table 1 is profoundly different from that observed for samples from previous Apollo missions since, prior to collection, the Taurus-Littrow samples had been subjected to bombardment by the intense series of solar flares of August 4–9, 1972. Large enhancements in the yields of 22Na, 40Sc, 54Mn, and 56Co are due to the solar event of August 1972.
The high intensity and energy of the flare made it possible for several groups to identify $^7$Be in a lunar sample for the first time. During the preliminary examination of the Apollo 17 samples O'Kelley et al., Rancitelli et al., and Schonfeld, all obtained results on $^7$Be concentrations of some samples by nondestructive gamma-ray spectrometry, and these data were published in LSPET (1973b). In addition, Finkel et al. (1973) reported measurements of $^7$Be abundance in Apollo 17 soils by use of destructive radiochemical analysis methods of high sensitivity.

Because chemical analysis data are lacking for most of the samples reported here, detailed interpretations cannot be made in some cases. However, the chemical analyses carried out during the preliminary examination of the Apollo 17 samples and reported by LSPET (1973a, 1973b) showed that the major element compositions of rocks and soils at Taurus-Littrow are similar to those at the

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**Table 1. Concentrations (dpm/kg) of cosmogenic radionuclides in Apollo 17 samples. Decay corrected to 2300 GMT, December 14, 1972.**

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Type*</th>
<th>Mass (g)</th>
<th>$^{214}$Al</th>
<th>$^{226}$Ra</th>
<th>$^{210}$Po</th>
<th>$^{40}$K</th>
<th>$^{40}$Ca</th>
<th>$^{137}$Cs</th>
<th>$^{109}$Cd</th>
<th>$^{13}$N</th>
<th>$^{7}$Be</th>
</tr>
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<tr>
<td>Rocks</td>
<td></td>
<td></td>
<td></td>
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<td></td>
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</tr>
<tr>
<td>70135</td>
<td>CB°</td>
<td>446</td>
<td>38 ± 2</td>
<td>33 ± 3</td>
<td>56 ± 6</td>
<td>56 ± 6</td>
<td>32 ± 3</td>
<td>10 ± 5</td>
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<tr>
<td>70183</td>
<td>CB</td>
<td>449</td>
<td>70 ± 4</td>
<td>50 ± 4</td>
<td>95 ± 10</td>
<td>105 ± 10</td>
<td>47 ± 5</td>
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<tr>
<td>71135</td>
<td>FR°</td>
<td>36.2</td>
<td>80 ± 6</td>
<td>95 ± 7</td>
<td>140 ± 15</td>
<td>290 ± 50</td>
<td>70 ± 30</td>
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<td>71136</td>
<td>FR°</td>
<td>25.8</td>
<td>90 ± 8</td>
<td>93 ± 9</td>
<td>160 ± 60</td>
<td>300 ± 70</td>
<td>70 ± 30</td>
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<td>71175</td>
<td>MB</td>
<td>188</td>
<td>60 ± 3</td>
<td>68 ± 4</td>
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<td>120 ± 30</td>
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<td>71566</td>
<td>CB</td>
<td>415</td>
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<td>49 ± 3</td>
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<tr>
<td>73215</td>
<td>BR</td>
<td>1057</td>
<td>59 ± 2</td>
<td>59 ± 5</td>
<td>36 ± 8</td>
<td>51 ± 20</td>
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<td>73355</td>
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<td>392</td>
<td>75 ± 4</td>
<td>88 ± 6</td>
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<td>430</td>
<td>107 ± 5</td>
<td>99 ± 6</td>
<td>78 ± 12</td>
<td>96 ± 20</td>
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<td>76295</td>
<td>BR°</td>
<td>261</td>
<td>67 ± 5</td>
<td>54 ± 4</td>
<td>38 ± 15</td>
<td>41 ± 7</td>
<td>5 ± 2</td>
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<td>78597</td>
<td>FB</td>
<td>319</td>
<td>48 ± 4</td>
<td>33 ± 4</td>
<td>80 ± 10</td>
<td>80 ± 20</td>
<td>25 ± 10</td>
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<td>CB</td>
<td>316</td>
<td>70 ± 3</td>
<td>63 ± 3</td>
<td>120 ± 12</td>
<td>153 ± 8</td>
<td>65 ± 3</td>
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<tr>
<td>Soils (&lt; 1 mm fines)</td>
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<td>71311</td>
<td>BS</td>
<td>50.1</td>
<td>69 ± 4</td>
<td>62 ± 6</td>
<td>102 ± 13</td>
<td></td>
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<tr>
<td>71501</td>
<td>BS</td>
<td>100</td>
<td>73 ± 4</td>
<td>89 ± 6</td>
<td>135 ± 15</td>
<td></td>
<td></td>
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<tr>
<td>73121</td>
<td>TT</td>
<td>100</td>
<td>159 ± 6</td>
<td>159 ± 9</td>
<td>137 ± 20</td>
<td>218 ± 20</td>
<td>15 ± 5</td>
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<td>73131</td>
<td>BS</td>
<td>100</td>
<td>54 ± 3</td>
<td>126 ± 5</td>
<td>75 ± 10</td>
<td>119 ± 12</td>
<td>15 ± 3</td>
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<tr>
<td>73411</td>
<td>TB</td>
<td>101</td>
<td>62 ± 3</td>
<td>49 ± 5</td>
<td>26 ± 8</td>
<td>20 ± 10</td>
<td>10 ± 5</td>
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<tr>
<td>73221</td>
<td>SS</td>
<td>46.3</td>
<td>197 ± 10</td>
<td>310 ± 15</td>
<td>230 ± 30</td>
<td>810 ± 40</td>
<td>33 ± 6</td>
<td>450 ± 350</td>
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<td>73241</td>
<td>TT</td>
<td>100</td>
<td>92 ± 5</td>
<td>110 ± 5</td>
<td>80 ± 8</td>
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<td>73261</td>
<td>TB</td>
<td>100</td>
<td>57 ± 4</td>
<td>42 ± 4</td>
<td>52 ± 12</td>
<td>5 ± 10</td>
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<tr>
<td>73281</td>
<td>TB</td>
<td>50.3</td>
<td>46 ± 5</td>
<td>42 ± 9</td>
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<td>76501</td>
<td>RS</td>
<td>97.9</td>
<td>90 ± 9</td>
<td>90 ± 8</td>
<td>60 ± 10</td>
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<td>78501</td>
<td>RS</td>
<td>113</td>
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<td>96 ± 10</td>
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<td>79231</td>
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<td>100</td>
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<tr>
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<td>TB</td>
<td>100</td>
<td>45 ± 4</td>
<td>43 ± 4</td>
<td>44 ± 6</td>
<td>26 ± 10</td>
<td>15 ± 4</td>
<td></td>
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</tbody>
</table>

* CB = coarse basalt; FB = fine basalt; MB = medium basalt; BR = breccia; BS = bulk soil; TT = trench top; TM = trench middle; TB = trench bottom; SS = skim soil; RS = rake soil.

°Fragment or chip sampled from large boulder.
Further, the variation in major element concentrations of specific types of geologic samples appears to be sufficiently regular to permit estimation of target element concentrations where needed.

Except for the Fra Mauro materials, surface samples from previous Apollo sites typically showed $^{26}$Al/$^{27}$Na concentration ratios $\approx 2$ for samples in which the $^{26}$Al concentration had reached its saturation value. Such concentration ratios have proved to be useful guides in searching for rocks with low exposure, since the $^{27}$Na concentration measures short-term exposure and the $^{26}$Al concentration determines cosmic ray exposure on a scale of a few million years. However, the ratio $^{26}$Al/$^{27}$Na in most Apollo 17 samples is close to unity because the intense solar flare bombardment in August 1972 more than doubled the amount of $^{27}$Na already present before the flare occurred. In addition, uncertainties in chemical composition of the samples make it difficult to identify samples of low exposure with respect to the 0.74 m.y. half-life of $^{26}$Al.

The elevated yields of $^{56}$Co and $^{55}$Mn over samples examined previously are due to solar proton bombardment of iron. As will be shown later, the $^{56}$Co concentration is especially useful in measuring the total proton flux, while $^{55}$Mn is a product of higher energy proton bombardment and is helpful in determining the rigidity of the proton energy distribution.

Similarly, the high yields of $^{46}$Sc in Apollo 17 surface samples arise from solar proton irradiation of titanium. Concentrations of $^{46}$Sc, corrected for galactic cosmic ray production in iron, correlate well with estimates and measurements of titanium target element concentrations in these samples. Although sixteen-day $^{51}$V is produced in solar flares by the reaction $^{46}$Ti($p$, $n$)$^{51}$V, over four months had elapsed between the August, 1972 solar flare and collection of the samples at Taurus-Littrow. Because of this long decay period following the solar flare, the concentrations of $^{51}$V reported in Table 1 correspond to the steady-state concentration of $^{51}$V expected (O'Kelley et al., 1972) for the bombardment of iron by high-energy, galactic protons.

Concentration patterns for some of the radionuclides discussed above suggest that some of the rocks listed in Table 1 were shielded at least partially from the most recent solar flare. Three possibilities are 73215, for which no orientation exists, 70185, which may have been buried, and 78597, a rake sample. Additionally, 70135 and 76295 are boulder chips which either were shadowed by overhanging boulder material as partial shielding, or were irradiated by a solar proton flux whose average energy was a function of the incident angle. The latter explanation based on solar proton anisotropy has been applied by Rancitelli et al. (1974) to explain the low ratio of $^{56}$Co to $^{55}$Mn in boulder chip 76295.

**Characterization of the August 1972 solar flare**

Chemical analysis data are lacking for many of the samples which might be used to characterize the solar flare of August, 1972. For this reason, $^{56}$Co and $^{55}$Mn concentrations may be used to advantage, since both are produced by solar proton bombardment of iron, and concentrations of iron in the Apollo 17 samples...
concerned may be estimated by analogy to samples of known composition to an accuracy of about ±10%. Excitation functions for production of $^{56}$Co and $^{54}$Mn are shown in Fig. 1 from the work of Brodzinski et al. (1971) and references therein. As shown in Fig. 1 the excitation function for production of $^{56}$Co from iron has an effective threshold of about 6 MeV; hence, the $^{56}$Co concentrations in thin surface samples is especially useful as a monitor of total proton flux. Information on the rigidity of the flare can be derived from the yields of $^{54}$Mn, a product of higher energy proton bombardment, with a threshold of about 25 MeV.

Data on solar flare production of $^{54}$Mn and $^{56}$Co in five thin surface samples are compiled in Table 2. Concentrations shown earlier in Table 1 were corrected for galactic cosmic ray production by methods discussed by O’Kelley et al. (1971), and then the solar production component was corrected for decay to August 7, 1972. The low-energy product $^{56}$Co exhibits very high concentration gradients at
exposed surfaces, so the concentrations of "Co determined for the thin surface samples of Table 2 are very high with respect to the high-energy product "Mn. This is especially striking in the case of skim soil 73221, which has an effective thickness of only about 0.6 g/cm² and a concentration ratio "Co/"Mn of 9.2.

The data were analyzed in terms of a model which for simplicity assumed the solar proton spectrum to be of the form

\[ \frac{dJ}{dE} = kE^{-\alpha} \]

where \( J \) is the proton flux, \( E \) is the proton energy in MeV, \( k \) is a constant related to the flare intensity, and \( \alpha \) is a shape parameter which measures the average energy of the flare. The model assumed the irradiated surface to be an infinite plane. The proton energy spectrum and relative flux were calculated as a function of depth, assuming that the number of secondary particles was negligible. Production of "Mn and "Co within each differential of thickness was obtained by integrating the product of the number of target atoms and the excitation function for the appropriate reaction over the attenuated energy spectrum. Details of more extensive calculations of this type have been discussed by Reedy and Arnold (1972) and by Rancitelli et al. (1971).

Results of the analysis are given in Table 3 and are compared with a report by Rancitelli et al. (1974) and a summary of satellite measurements compiled by King (1973). Agreement between all groups is quite satisfactory, even though the first two measurements of Table 3 were carried out on lunar surface samples and the data compiled by King (1973) were obtained from satellite measurements in a relatively unshielded region of space. The very high value obtained for the integrated proton flux \( J_{1}(> 10 \text{ MeV}) = 1.9 \times 10^{10} \text{protons/cm}^2\text{ }\) verified that the solar flare event of August 4-9, 1972 was the largest of solar cycle 20. In fact, the solar proton fluences observed for the period August 4-9, 1972 constitute about 70% of the proton fluence above 10 MeV for the entire cycle (King, 1973). A shape parameter \( \alpha = 1.9 \) also implies that the average energy, or "rigidity," of the protons from the August 1972 flare is much higher than the average energy of the present solar cycle, which is characterized by a shape parameter \( \alpha = 3.0 \).
Cosmogenic radionuclides in samples from Taurus-Littrow

Table 3. Integrated proton flux \( J(E > 10 \text{ MeV}) \) and spectrum data for the August 4-9, 1972 solar flare.

<table>
<thead>
<tr>
<th>( J(E &gt; 10 \text{ MeV}) ) protons/cm(^2) year</th>
<th>Shape parameter, ( \alpha )</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 1.9 \times 10^{10} )</td>
<td>1.9 ( \pm 0.2 )</td>
<td>Present work</td>
</tr>
<tr>
<td>( 1.58 \times 10^{10} )</td>
<td>2.0 ( \pm 0.1 )</td>
<td>Rancitelli et al. (1974)</td>
</tr>
<tr>
<td>( 2.25 \times 10^{9} )</td>
<td>-</td>
<td>King (1973)</td>
</tr>
</tbody>
</table>

*The parameter \( \alpha \) in the energy distribution \( dJ/dE \propto E^\alpha \).*

Trench samples

Of thirteen soil samples from the valley of Taurus-Littrow which we have analyzed, more than half were associated with trenching operations. Because samples from a trench give a rather low-resolution view of the variations in radionuclide concentration with depth, as compared with that obtained from segments of a core in the lunar regolith, our discussion will be rather qualitative. However, trench samples are of much more interest for their information on color, texture, mineralogy, and chemistry (Eldridge et al., 1974).

Sample 73121 was discussed earlier, and sample 73141 was taken from a trench near 73121 at a depth of 15 cm in the light mantle. The radionuclide concentrations found at 15 cm depth and shown in Table 1 are those expected for galactic cosmic ray production. Not related to this trench, as might be expected from number, is 73131. This sample was removed from the wall of a 2 m diameter crater, 15 cm down the crater wall and may contain particles from the comminution of a soil clod or soft breccia removed simultaneously with the soil. Sample 73131 has received an intense, recent, solar-flare irradiation; however, the concentration of \(^{26}\)Al appears to be well below the surface saturation value. Although speculative in the absence of more detailed information concerning the sampling procedures used, the low \(^{26}\)Al content of 73131 suggests that the small 2 m crater at Station 2a was formed within the last million years.

From Station 3 we obtained a skim soil 73221, light-gray soil 73241 from 0 to 5 cm depth of a trench, a medium-gray soil 73261 from 5 to 10 cm depth, and a light-gray soil 73281 from an adjacent zone in the trench, also from about 5 to 10 cm depth. Again, the expected depth dependence was found, as shown in Table 1. It will be noted that for our present purpose, 73261 and 73281 are duplicate samples and exhibit essentially the same radionuclide concentrations.

At Station 9, on the southeast flank of the Van Serg Crater ejecta blanket, sample 79221 was taken from the uppermost 2 cm of a trench. Data from this sample were used earlier in Table 2. Sample 79261, from a sampling depth of 7 to 17 cm, was removed from a light-gray or white layer. The expected decrease in radionuclide concentrations from surface to depth again was noted. The concentrations of \(^{26}\)Al, \(^{22}\)Na, \(^{40}\)Mn in 79261 agree well with calculations of galactic cosmic ray production rates (cf. Reedy and Arnold, 1972), but concentrations of \(^{54}\)Co and \(^{40}\)Sc are somewhat higher than expected.
Radionuclides produced by solar proton bombardment, especially $^{60}$Co, are sensitive indicators of the mixing of surface soil into deeper trench samples. Data on $^{60}$Co presented in Table 1 demonstrate that the trenching discussed in this section was carried out under excellent control. Only 79261 may show evidence for material mixed from above.

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, R. S. Clark, and M. A. Reynolds, for their aid in rapidly and efficiently processing the samples for this study.

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LSPET (Lunar Sample Preliminary Examination Team) (1973a) Apollo 17 lunar samples: Chemical and petrographic description. Science 182, 659-672.


Primordial and cosmogenic radionuclides in Descartes and Taurus-Littrow materials: Extension of studies by nondestructive gamma-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 Apollo 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 to 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.

*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.
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 ~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 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 gamma-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 gamma-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 Figure 1. Two unsaved 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 ~20-30% lower in radioelement content than the average of 11 soils from 8 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 came 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 to 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 (Eloridge 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) de-
scribed 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 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 micro-
probe analyses of small samples from these inhomogeneous rocks yielded K con-
centrations 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 con-
centrations 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 simi-
larity 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 -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 in Figure 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 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 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 2.6, 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 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, 1970b and 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 Apollo 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 labelled 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 the extreme differentiation in lunar crustal materials. The primordial radioelement relationships may be coupled with past and future orbital gamma-ray and x-ray experiments to delineate the crustal distributions of these important materials.
Cosmogenic radionuclides

The 2-1/2 years 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-year $^{22}$Na and $7.4 \times 10^5$-year $^{26}$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 that the concentration of $3.7 \times 10^6$-year $^{53}$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 years) makes it a useful indicator of recent solar cosmic-ray exposure, while $^{26}$Al ($7.4 \times 10^5$ years) 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 grams, appears to be saturated in $^{26}$Al, while 67785, 5.0 grams, 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 radio-nuclide 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.
REFERENCES


Table 1. Apollo 16 radioelement concentrations.

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>Type</th>
<th>K (ppm)</th>
<th>Th (ppm)</th>
<th>U (ppm)</th>
<th>Th/U</th>
<th>K/U</th>
</tr>
</thead>
<tbody>
<tr>
<td>8 Sta Avg *</td>
<td>&lt;1 mm fines</td>
<td>940 ± 50</td>
<td>1.95 ± .09</td>
<td>0.54 ± .03</td>
<td>3.61 ± .26</td>
<td>1740 ± 130</td>
</tr>
<tr>
<td>63501 *</td>
<td>&lt;1 mm fines</td>
<td>728 ± 50</td>
<td>1.53 ± .08</td>
<td>0.41 ± .03</td>
<td>3.73 ± .34</td>
<td>1780 ± 180</td>
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<tr>
<td>63320</td>
<td>Unsieved fines</td>
<td>800 ± 40</td>
<td>1.35 ± .07</td>
<td>0.39 ± .03</td>
<td>3.46 ± .32</td>
<td>2050 ± 190</td>
</tr>
<tr>
<td>63340</td>
<td>Unsieved fines</td>
<td>790 ± 40</td>
<td>1.33 ± .07</td>
<td>0.40 ± .03</td>
<td>3.33 ± .30</td>
<td>1980 ± 180</td>
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<tr>
<td>67016,2</td>
<td>Breccia</td>
<td>485 ± 25</td>
<td>0.69 ± .03</td>
<td>0.20 ± .01</td>
<td>3.45 ± .23</td>
<td>2420 ± 180</td>
</tr>
<tr>
<td>67115,9</td>
<td>Breccia</td>
<td>475 ± 25</td>
<td>0.44 ± .02</td>
<td>0.12 ± .01</td>
<td>3.67 ± .35</td>
<td>3960 ± 390</td>
</tr>
<tr>
<td>67055 *</td>
<td>Breccia</td>
<td>1620 ± 80</td>
<td>3.69 ± .18</td>
<td>0.98 ± .05</td>
<td>3.77 ± .27</td>
<td>1650 ± 120</td>
</tr>
<tr>
<td>67937 *</td>
<td>Breccia</td>
<td>1650 ± 90</td>
<td>3.24 ± .16</td>
<td>0.96 ± .05</td>
<td>3.38 ± .24</td>
<td>1720 ± 130</td>
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<tr>
<td>60618,0</td>
<td>Crystalline</td>
<td>670 ± 50</td>
<td>0.63 ± .06</td>
<td>0.28 ± .03</td>
<td>2.25 ± .32</td>
<td>2390 ± 310</td>
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<tr>
<td>65785,0</td>
<td>Crystalline</td>
<td>1850 ± 150</td>
<td>3.03 ± .18</td>
<td>0.97 ± .07</td>
<td>3.12 ± .29</td>
<td>1970 ± 210</td>
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<tr>
<td>62295,0 *</td>
<td>Crystalline</td>
<td>630 ± 30</td>
<td>3.20 ± .15</td>
<td>0.82 ± .05</td>
<td>3.90 ± .30</td>
<td>770 ± 60</td>
</tr>
</tbody>
</table>

*Data previously described in Eldridge et al., (1973).
Table 2. Apollo 17 radioelement concentrations.

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>Type</th>
<th>K (ppm)</th>
<th>Th (ppm)</th>
<th>U (ppm)</th>
<th>Th/U</th>
<th>K/U</th>
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<tr>
<td>6 Sple. Avg. *</td>
<td>F. basalt</td>
<td>382</td>
<td>0.40</td>
<td>0.14</td>
<td>3.00 ± .26</td>
<td>2930 ± 260</td>
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<tr>
<td>74245,0</td>
<td>F. basalt</td>
<td>620 ± 30</td>
<td>0.40 ± .03</td>
<td>0.13 ± .02</td>
<td>3.08 ± .53</td>
<td>4770 ± 770</td>
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<tr>
<td>71546,0</td>
<td>F. basalt</td>
<td>500 ± 25</td>
<td>0.40 ± .03</td>
<td>0.15 ± .02</td>
<td>2.67 ± .41</td>
<td>3333 ± 475</td>
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<tr>
<td>4 Sple. Avg. *</td>
<td>M. basalt</td>
<td>444</td>
<td>0.38</td>
<td>0.11</td>
<td>3.48 ± .24</td>
<td>4015 ± 240</td>
</tr>
<tr>
<td>72155,1</td>
<td>M. basalt</td>
<td>525 ± 25</td>
<td>0.36 ± .02</td>
<td>0.13 ± .01</td>
<td>2.77 ± .26</td>
<td>4040 ± 365</td>
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<td>6 Sple. Avg. *</td>
<td>C. basalt</td>
<td>495</td>
<td>0.34</td>
<td>0.11</td>
<td>3.12 ± .16</td>
<td>4590 ± 250</td>
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<tr>
<td>70315,0</td>
<td>C. basalt</td>
<td>400 ± 20</td>
<td>0.27 ± .02</td>
<td>0.10 ± .01</td>
<td>2.70 ± .34</td>
<td>4000 ± 450</td>
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<td>&lt; 1 mm fines</td>
<td>595 ± 30</td>
<td>0.73 ± .07</td>
<td>0.26 ± .03</td>
<td>2.81 ± .42</td>
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<td>72161,26</td>
<td>&lt; 1 mm fines</td>
<td>795 ± 40</td>
<td>1.47 ± .08</td>
<td>0.45 ± .04</td>
<td>3.27 ± .34</td>
<td>1770 ± 180</td>
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*Eldridge et al. (1974).
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<th>Sample Number</th>
<th>Mass (g)</th>
<th>$^{22}\text{Na}$</th>
<th>$^{26}\text{Al}$</th>
<th>$^{54}\text{Mn}$</th>
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<td>20.5</td>
<td>45 ± 10</td>
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<td>63340,18</td>
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<td>91 ± 4</td>
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<tr>
<td>65785,0</td>
<td>5.0</td>
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<td>59 ± 6</td>
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<td>88 ± 4</td>
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<td>67 ± 3</td>
<td>30 ± 15</td>
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<td><strong>Taurus-Littrow</strong></td>
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*Concentrations of $^{22}\text{Na}$ and $^{54}\text{Mn}$ corrected for decay to 23 April 1972 and 14 December 1972 for Apollo 16 and 17 samples, respectively.
CAPTIONS TO FIGURES

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).

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).

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).

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 end members.
Fig. 4