

cific cases it may be possible by arranging proper combination of phases, e.g., basaltic liquid, pyroxene, and spinel, to obtain a situation where the liquid REE abundances are unfractionated with respect to the bulk solid. This is, however, a special case that would probably not produce the other element abundances and ratios, e.g., Ti, Zr, Al, and Cr (1), observed in the Tranquillity Base basalts. It has been proposed that the relative lack of REE fractionation in the ORB is due to complete extraction of the REE from the earth's mantle (4). The low Al content and high Ti content found for the lunar materials is quite inconsistent with this hypothesis for the lunar case. A detailed study of the ORB shows that the total REE, FeO, and TiO<sub>2</sub> contents increase together and, furthermore, that those rocks high in total REE have small negative Eu anomalies (5). The explanation proposed for these variations in the ORB abundance patterns is the removal of basic plagioclase and olivine (5). A similar hypothesis is proposed here to explain the abundance patterns observed in the lunar basalts. The original composition and pattern of a liquid consisting of 6 parts of plagioclase (An 96), 1½ parts olivine (Fo 80) and 2½ parts of "coarse grained" liquid (10062 and 10003 averaged) are shown in Fig. 1. We recognize that absence of pyroxene from such a liquid may be unusual. If pyroxene were removed from the parent liquid in significant amounts, less than 10 percent, the Cr content of this initial liquid would have to be over 1 percent by weight (4). Assuming that the distribution coefficients between liquid and plagioclase for Eu, Sr, Ba, and REE are 1.3, 0.7, 10, and 20 (2), respectively, the CaO, Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, FeO, and MgO concentrations of such an original liquid would be about 15, 25, 3, 7½, and 8 percent, respectively. If this hypothesis is correct, we infer that cumulates rich in plagioclase should occur on the lunar surface. It is also of some interest that the results of the Surveyor 7 analysis (6) indicate unusually high Al<sub>2</sub>O<sub>3</sub> (21 percent) and CaO (15 percent) contents and relatively low Fe contents are found in highland regions.

The depletion of Ba and La relative to heavy REE and broad peak at Sm and Gd is not explained by this process. These features could be due to other minor phases that have crystallized and separated from the liquid or to fractionation that takes place during production of parent liquid in the lunar interior. If the proposed original liquid is pro-

duced by partial melting in a lunar mantle (< 20 percent) we infer that the U, Th, and K contents of the parent lunar mantle are less than 0.08, 0.3 and 100 ppm, respectively.

The Sr<sup>87</sup>/Sr<sup>86</sup> ratios and Rb<sup>87</sup>/Sr<sup>86</sup> ratios of six igneous rocks are shown in an isochron diagram in Fig. 2. With the exception of sample 10017, these data define an isochron indicating that the time of crystallization was 3.5 ± 0.3 billion years ago. We have no explanation for sample 10017 but note again that our sample was a surface chip. The limits placed on the difference between the initial ratio (0.6991) and the achondrite initial ratio, 0.6989 (7), indicate that the source of these igneous rocks has a rubidium-strontium ratio less than one-tenth that of chondrites. If the igneous liquids were derived from the interior of the moon 3.5 ± 0.3 billion years ago, we conclude that large portions of the moon must be even more depleted in rubidium relative to strontium than the earth (8). The possibility that the high abundance of refractory elements e.g., Ca, Al, TiO<sub>2</sub>, REE, and so forth, is a primary characteristic of a large part

of the moon should not be overlooked. Unless this possibility is excluded one cannot infer general differentiation of the moon from the existence of basaltic rocks on the lunar surface.

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### Rare Earth Elements in Returned Lunar Samples

**Abstract.** *A linear correlation between concentrations of Sm and ratios of Sm to Eu for nine lunar samples suggests that those samples could correspond to liquids from equilibrium partial melting of a common source. On the basis of partition coefficients in terrestrial systems, the fraction of melting would not have exceeded about 15 percent and the immediate source could have been composed of olivine, orthopyroxene, and opaque minerals plus at least 25 percent feldspar, with at most a few percent calcic clinopyroxene and less than 1 percent apatite. The large Eu depletions could also have been produced by fractional crystallization if the ratio of Eu<sup>2+</sup> to Eu<sup>3+</sup> in lunar magmas significantly exceeds the values for terrestrial magmas.*

We present here concentrations for 12 of the rare earth elements (REE) in nine samples of lunar material returned to earth by the Apollo 11 astronauts (1). Included are five fine-grained igneous rocks (type A), one medium-grained igneous rock (type B), two breccias (type C), and "fine" material less than 1 mm in diameter (type D) from the lunar soil. The igneous rocks were received as 1-g chips, and the breccias and soil as cuts of powders. The concentrations of the REE were measured by neutron activation analysis; the REE were separated as a group from the irradiated samples and then were assayed by Ge-Li gamma-ray spectrometry.

The REE concentrations for the lunar samples plus those for a terrestrial basalt

which was analyzed in the same irradiation as the lunar breccias are given in Table 1. The accompanying uncertainties are based strictly on counting statistics and are applicable to the comparison of one REE with the others in the same rock. Values for all the REE in any particular sample may be systematically in error by 1 to 2 percent.

Relative and absolute REE abundances for these samples are compared in Fig. 1 with those for a composite of nine chondritic meteorites (2). The ratios of the concentrations in parts per million for each element in a sample to the concentration in parts per million for the same element in the chondrites are plotted as a function of REE atomic number (3). Included as a dashed line

Table 1. Rare earth concentrations in parts per million for nine lunar materials and an oceanic andesite\*.

Element	Sample number and type									EPR-D-3 basalt
	10022 A	10057 A	10072 A	10003 B	10020 B	10045 B	10048 C	10060 C	10084 D	
La	26.4 ± 0.7	28.2 ± 0.4	22.7 ± 0.5	14.1 ± 0.2	8.4 ± 0.2	6.7 ± 0.3	20.2 ± 0.3	20.8 ± 0.4	16.6 ± 0.4	20.4 ± 0.4
Ce	68 ± 2	75 ± 1	69 ± 1	41.3 ± 0.7	25.6 ± 0.6	22.5 ± 0.4	56 ± 1	58 ± 1	47 ± 1	61 ± 1
Nd	66 ± 4	69 ± 4	51 ± 4	42.5 ± 3	31 ± 3	21 ± 2	41 ± 7	46 ± 15	43 ± 6	57 ± 10
Sm	21.2 ± 0.1	20.8 ± 0.1	17.9 ± 0.2	13.1 ± 0.1	9.94 ± 0.06	7.94 ± 0.05	14.8 ± 0.2	15.4 ± 0.1	13.7 ± 0.1	15.9 ± 0.1
Eu	2.04 ± 0.01	2.18 ± 0.02	2.07 ± 0.04	1.80 ± 0.01	1.75 ± 0.01	1.52 ± 0.03	1.95 ± 0.02	2.06 ± 0.02	1.77 ± 0.02	4.05 ± 0.03
Gd	25 ± 2	26 ± 3	26 ± 2	17 ± 2	16 ± 2	12.3 ± 0.9	20 ± 1	24 ± 2	15 ± 2	22 ± 2
Tb	4.7 ± 0.1	5.0 ± 0.1	4.3 ± 0.1	3.26 ± 0.07	2.59 ± 0.06	2.11 ± 0.06	3.5 ± 0.1	3.6 ± 0.1	3.2 ± 0.2	3.6 ± 0.1
Dy	31.2 ± 0.3	34.7 ± 0.2	31.2 ± 0.3	22.4 ± 0.3	17.8 ± 0.1	14.4 ± 0.1	25.0 ± 0.3	26.3 ± 0.2	20.1 ± 0.1	25.1 ± 0.2
Ho	5.5 - 8.7	5.8 - 7.0	5.2 - 8.4	3.7 - 4.4	3.0 - 3.6	2.2 - 3.5	4.2 - 5.2	4.4 - 5.4	4.2 - 4.7	4.5 - 5.5
Er	16 ± 3	19 ± 2	16 ± 3	12 ± 1	9 ± 1	8.7 ± 2	14 ± 4	16 ± 5	12 ± 2	19 ± 6
Yb	17.7 ± 0.5	18.8 ± 0.2	16.6 ± 0.5	11.9 ± 0.3	9.8 ± 0.2	8.6 ± 0.3	12.5 ± 0.3	13.2 ± 0.3	11.3 ± 0.3	14.8 ± 0.3
Lu	2.55 ± 0.02	2.66 ± 0.02	2.24 ± 0.01	1.69 ± 0.01	1.41 ± 0.01	1.17 ± 0.01	2.10 ± 0.02	2.00 ± 0.02	1.66 ± 0.01	2.29 ± 0.02

\*This rock was classified as a basalt in reference 4 but is now known to be an andesite (S. Hart, private communication).

in the upper part of Fig. 1 are the ratios relative to chondrites of a composite of shales (2). The REE abundances of these shales are typical of surface matter from the terrestrial continents.

The curves for rocks 10057 and 10045 (Fig. 1) represent the extremes of REE concentrations among our nine samples. The REE concentrations for the soil, which presumably represents some kind of average for the area around Tranquillity Base, fall between the extremes and are about 10 percent lower for the lighter REE and 20 percent lower for the heavier REE than the average for the other eight samples. Except for Eu, the relative abundances of the

REE in these lunar samples are rather similar to those in the chondrites, in sharp contrast to terrestrial continental matter. Relative to the other REE, and with respect to chondrites, Eu is much more depleted (0.25 to 0.5 times the interpolated values from Fig. 1) in these lunar materials than in the shales (0.67 times). The similarity in REE abundances (except for Eu) between the two lunar breccias and the basalt from the East Pacific Rise (lower part of Fig. 1) is remarkable.

There is no compelling reason to presume that the REE abundances in the samples we analyzed are representative of the entire Sea of Tranquillity or of

lunar maria in general. The maria comprise about one-fifth of the lunar surface. If the average abundance of a REE, for example, Sm, in the whole moon is 0.18 ppm as in the chondrites, then at an average density of 3.3 g/cm<sup>3</sup>, for every kilometer of depth, the mare could contain as much as 15 percent of all the lunar Sm.

The similarity between the REE abundances in these lunar samples and those in the oceanic andesite does not suggest that the moon came from the Pacific Ocean basin. The sample chosen for comparison, EPR-D-3, is three times richer in REE than most other subalkaline basalts from the ocean ridges and has a significant Eu anomaly which most of these basalts do not have (4). There are significant differences in bulk composition (1, 4). Calcium-rich achondrites also have relative REE abundances similar to those in chondrites but the absolute REE abundances are several times greater (5). We suggest that similar processes produced the similar REE abundances in the oceanic and lunar rocks. Both types of rocks are igneous, most are fine-grained, and they presumably crystallized fairly rapidly from silicate melts (1).

Processes which act on or generate silicate melts and which can reasonably be expected to alter REE abundances in them are fractional crystallization and partial melting. We consider here only the problem of generating by those processes the variations in abundances of Eu relative to Sm and Gd in these lunar rocks. We presume also that the relative REE abundances found in chondrites are those of the starting material for objects native to the solar system (5) and would like to account for the Eu anomalies relative to chondrites by those processes.

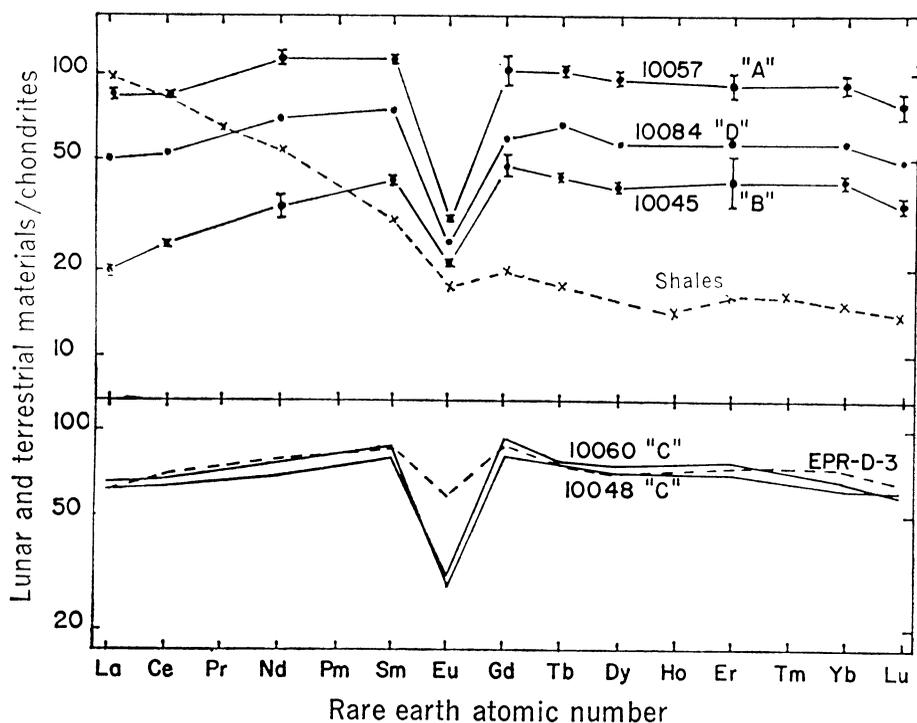


Fig. 1. Comparison diagram (2, 3) for lunar materials (solid lines), a composite of North American shales (2) (upper dashed line), and a submarine andesite (4) from the East Pacific Rise (lower dashed line).

It is convenient for this purpose to use the ratios of concentrations of Sm to Eu. From Figs. 1 and 2 it can be seen that the ratios of Sm to Eu increase as the absolute abundances of Sm and the other REE in the rocks increase. Europium is taken up in preference to the other REE by feldspars, presumably through its reduction to the 2+ oxidation state (5, 6). When minerals separated from a basic rock are analyzed, most of the Eu is usually found in the feldspar; the bulk of the other REE is found in clinopyroxenes and in apatite and other minor REE-concentrating minerals when they are present. Europium has not been found to be preferentially concentrated in any other mineral. Crystallization of feldspar from a silicate liquid should therefore give rise to a smooth variation of the ratio of Sm and Eu in the residual liquid. Alternatively, partial melting to different extents of a rock containing feldspar should yield liquids with smoothly varying ratios of Sm and Eu.

Mathematical models for the behavior of trace elements during these processes have been developed (7). Their use depends on knowledge of partition coefficients for those elements between the liquid and solid phases present. Partition coefficients,  $D$ , taken here to be the ratio of the concentration of the element in a solid phase to its concentration in the liquid, have been obtained by analysis of phenocrysts and host matrices, treatment of associated gabbros and ultramafics as melt-residue pairs, analyses of minerals in the Skaergaard and Kiglapait intrusions, and laboratory studies of synthetic systems (8). Some variations have been observed, but the general agreement among these several methods indicates that the values of interest can be used with confidence, at least for common terrestrial silicate liquids. We have used the following values in our calculations;  $D(\text{Eu}, \text{plagioclase})$ , 0.35;  $D(\text{Sm}, \text{plagioclase})$ , 0.03;  $D(\text{Eu}$  and  $\text{Sm}, \text{clinopyroxene})$ , 0.4;  $D(\text{Eu}$  and  $\text{Sm}, \text{apatite})$ , 10;  $D(\text{Eu}$  and  $\text{Sm}, \text{olivine}, \text{orthopyroxene}, \text{and opaque minerals})$ ,  $< 0.01$ .

During fractional crystallization, when the fraction of a system with average Sm content  $C_{\text{Sm},A}$  that has crystallized is  $x$ , the concentration of Sm in the residual liquid  $C_{\text{Sm},L}$  is

$$C_{\text{Sm},L} = C_{\text{Sm},A} (1 - x)^{D(\text{Sm})-1} \quad (1)$$

The ratio of the concentration of Sm to that of Eu in the system is

$$\frac{C_{\text{Sm},L}/C_{\text{Eu},L}}{(C_{\text{Sm},A}/C_{\text{Eu},A}) (1 - x)^{D(\text{Sm})-D(\text{Eu})}} \quad (2)$$

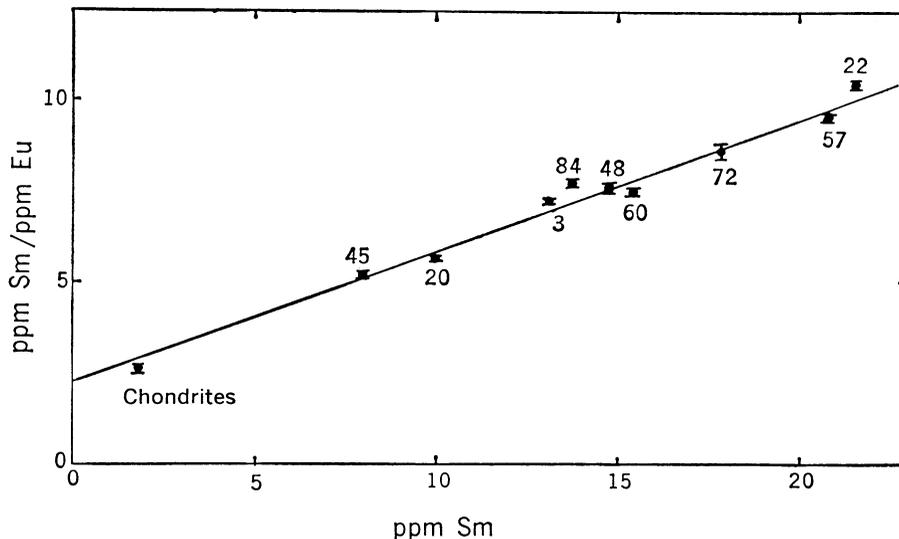


Fig. 2. Values for the ratios of Sm to Eu in the lunar samples vary linearly with concentrations of Sm. The line was obtained by a least-squares analysis of the nine points for the lunar samples.

The extent of separation between Eu and Sm depends on the difference between the partition coefficients. If we take a value for  $D(\text{Eu}, \text{plagioclase})$  of 0.35, the maximum published value, and a value for  $D(\text{Sm}, \text{plagioclase})$  of 0.03, the minimum conceivable value, and begin with a melt which has the same ratio of Sm to Eu as chondrites, 97 percent of the melt would have to crystallize as feldspar to produce the ratio found in the lunar soil, 88 percent of the melt would have to crystallize as feldspar to produce the ratio of rock 10045, and 99 percent of the melt would have to crystallize as feldspar to produce the ratio of rock 10022. This is clearly impossible. Simultaneous crystallization of other minerals would increase the necessary fraction crystallized, since the difference between the partition coefficients would be decreased.

Could the lunar rocks represent residual liquids from a system with an initial ratio of Sm to Eu lower than that of chondrites? This idea not only leaves unsolved the problem of generating such a ratio of Sm to Eu in the starting material, but also can be ruled out on the basis of the following argument. Elimination of  $x$  between Eq. 1 for Sm and the corresponding equation for Eu gives Eq. 3, which is linear in  $\ln C_{\text{Sm},L}/C_{\text{Eu},L}$  and  $\ln C_{\text{Sm},A}$ .

$$\ln \frac{C_{\text{Sm},L}}{C_{\text{Eu},L}} = \frac{D(\text{Sm}) - D(\text{Eu})}{D(\text{Sm}) - 1} \ln C_{\text{Sm},A} + \ln \left[ \frac{C_{\text{Sm},A}}{C_{\text{Eu},A}} \right]^{[D(\text{Eu})-1]/[D(\text{Sm})-1]} \quad (3)$$

A plot of  $\ln (C_{\text{Sm},L}/C_{\text{Eu},L})$  as a function of  $\ln (C_{\text{Sm},A})$  (not shown) is

approximately linear. The slope, as determined by least-squares analysis, is 0.70; this requires that the value for  $D(\text{Eu})$  exceed 0.71 for  $D(\text{Sm}) > 0$ , which is twice as high as the value allowed from measurements of partition coefficients. On the basis of the above partition coefficients, the lunar rocks would not appear to be solidified residues from fractional crystallization. If lunar magmas are sufficiently reducing, however, that their ratio of free ion activity of  $\text{Eu}^{2+}$  to  $\text{Eu}^{3+}$  exceeds the ratio for terrestrial magmas, the variations in ratios of Sm to Eu might still be explained by fractional crystallization.

To test the possibility that the lunar samples might correspond to different extents of partial melting of a common starting material with a ratio of Sm to Eu equal to that in chondrites, we have made several calculations using the formulation of Gast (7). Partial melting of a rock consisting of 50 percent feldspar and 50 percent orthopyroxene, with equal amounts of the two minerals forming each increment of the melt, provides the ratio of Sm to Eu in rock 10022 in 4 to 5 percent partial melting, and that in rock 10045 in 15 percent partial melting. Reducing  $D(\text{Eu}, \text{plagioclase})$  to 0.20 still makes it possible to produce the ratios in rocks 10022 and 10045 in less than 1 percent and 7 percent partial melting. Both models yield straight lines when plots like Fig. 2 are made.

The maximum ratio of Sm to Eu, relative to the ratio in the starting material, that can be obtained (by the minimum of partial melting) is equal to the ratio

of the partition coefficients between the liquid and the bulk rock, which varies according to the constituent minerals and their proportions. As a consequence, models that contained less than 25 percent of plagioclase, or more than a few percent of clinopyroxene, or as much as 1 percent of apatite, failed.

The linear relationship of Fig. 2 does not correspond to simple mixing of two materials with different ratios of Sm to Eu; such mixtures generate parabolic curves. This may explain why the values for the soil and one of the breccias lie off the line. Such mixing also excludes any significant contribution from non-equilibrium partial melting of homogeneous minerals.

If the lunar rocks represent partial melts from a starting material composed of feldspar and mafic constituents such as olivine, orthopyroxene, and opaque oxides, the absolute contents of the REE and the magnitudes of the ratios of Sm to Eu should be inversely correlated with the relative amount of felsic components in the rocks. Comparison of our results with analyses for the major constituents (1) indicates that such is the case.

Thus we conclude that these lunar rocks may very well represent liquids from different stages of equilibrium partial melting from a common starting material which could have had a ratio of Sm to Eu equal to that in chondrites. The very limited deviation of the points from the least-squares line in Fig. 2 indicates that the compositions of these liquids were not appreciably modified by fractional crystallization before freezing.

Those models that were successful in producing the requisite ratios of Sm to Eu in the lunar rocks from that in chondrites failed by a factor of 2 to 3 to concentrate those elements as much as was required in order that the starting material have chondritic absolute abundances. That plus the requirement of a high content of plagioclase in the starting material suggest that the bulk composition of the moon more closely resembles that of basalts (or basaltic achondrites) than that of chondrites, or that, in analogy with the upper mantle of the earth (9), the components of plagioclase and the REE were strongly concentrated toward the exterior of the moon.

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## Trace Elements and Radioactivity in Lunar Rocks: Implications for Meteorite Infall, Solar-Wind Flux, and Formation Conditions of Moon

**Abstract.** Lunar soil and type C breccias are enriched 3- to 100-fold in Ir, Au, Zn, Cd, Ag, Br, Bi, and Tl, relative to type A, B rocks. Smaller enrichments were found for Co, Cu, Ga, Pd, Rb, and Cs. The solar wind at present intensity can account for only 3 percent of this enrichment; an upper limit to the average proton flux during the last  $4.5 \times 10^9$  years thus is  $8 \times 10^9 \text{ cm}^{-2} \text{ yr}^{-1}$ . The remaining enrichment seems to be due to a 1.5 to 2 percent admixture of carbonaceous-chondrite-like material, corresponding to an average influx rate of meteoritic and cometary matter of  $2.9 \times 10^{-9} \text{ g cm}^{-2} \text{ yr}^{-1}$  at Tranquility Base. This is about one-quarter the terrestrial rate. Type A, B rocks are depleted 10- to 100-fold in Ag, Au, Zn, Cd, In, Tl, and Bi, relative to terrestrial basalts. This suggests loss by high-temperature volatilization, before or after accretion of the moon. Positron activities due mainly to  $^{22}\text{Na}$  and  $^{26}\text{Al}$  range from 90 to 220  $\beta^+ \text{ min}^{-1} \text{ kg}^{-1}$  in five small rocks or fragments (9 to 29 g). The higher activities presumably indicate surface locations. Th and U contents generally agree with those found by the preliminary examination team.

In a series of experiments, carried out by Keays, Ganapathy, Laul, and Anders, 12 lunar samples of types A to D and four samples of terrestrial rocks were analyzed for 16 trace elements by a radiochemical neutron activation procedure (1). Sample weights were typically  $\sim 0.1$  g; the integrated neutron flux was  $1 \times 10^{20} \text{ cm}^{-2}$  (Table 1).

The reproducibility of the method is indicated by triplicate analyses of lunar soil 84,49 (the third analysis, 84,49W, actually is a weighted mean of three individual analyses on sieve fractions, 84,49,1-3) and duplicates of Columbia River basalt BCR-1. Radiochemical purity was checked with  $\gamma$ -spectrum and half-life, in the case of a few Tl samples also by recycling. The absolute accuracy seems to be better than 10 percent for homogeneously distributed elements, judging from published analyses of basalt BCR-1.

Italicized values are considered meaningless or doubtful. They include all In and two Ag analyses on lunar samples, which were contaminated up to 10,000-fold the intrinsic level by In-Ag vacuum

gaskets on sample return containers. One of our Ir values was surprisingly high, probably due to laboratory contamination. No data were obtained on Te, because of severe interference from fission-produced  $^{131}\text{Te}$ - $^{131}\text{I}$ . The Pd numbers (determined via  $^{111}\text{Ag}$ ) are somewhat uncertain, because  $^{111}\text{Ag}$  is also made from  $^{109}\text{Ag}$  by double-neutron capture. Corrections for this effect were rather large, and not too well determined:  $\sim 10$  percent for  $\text{Ag}/\text{Pd} = 0.5$  and  $\sim 60$  percent for  $\text{Ag}/\text{Pd} = 5$ . Sample 47,32 (a rock powder prepared at the LRL) gave higher values for 7 elements than did any other A or B sample. We suspect that this was due to contamination during crushing, and therefore did not use any of the data for this sample.

The first 9 elements in Table 1 are 2 to 100 times more abundant in type C and D material than in average A, B rock (last 3 lines of Table 1). Inasmuch as C (breccia) and D (soil) probably are largely derived from A, B rock, it seems likely that the enrichment is due to addition of extraneous material: mete-