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 nonequilibrium 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* 4.5×10^9 *years thus is* 8×10^9 *cm*$^{-2}$ *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* 2.9×10^{-9} *g cm*$^{-2}$ *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}*Na and* ^{26}*Al range from 90 to 220* β^+ *min*$^{-1}$ *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 ~0.1 g; the integrated neutron flux was 1×10^{20} 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 γ-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 BRC-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}Te-^{131}I. The Pd-numbers (determined via ^{111}Ag) are somewhat uncertain, because ^{111}Ag is also made from ^{109}Ag by double-neutron capture. Corrections for this effect were rather large, and not too well determined: ~10 percent for Ag/Pd = 0.5 and ~60 percent for Ag/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: meteoritic, cometary, and solar wind. (Other factors may also play a role: mechanical sorting, volatilization by shock melting, and so forth. It is difficult to correct for them, though; and the overall consistency of our results for elements greatly differing in geochemical character such as Ir, Br, and Bi suggests that these effects are not large.)

In order to characterize this extraneous component, we have plotted in Fig. 1 the "excess" of each element in type C and D material over that in average A, B rock. The data were normalized to type I carbonaceous chondrites (solid line). This approach had previously been used by Mazor and Anders (*2*) in a study of gas-rich meteorites.

It is striking that the data for most elements lie parallel to the carbonaceous chondrite line, at an abundance of 0.01 to 0.02. Insofar as these elements are concerned, types C and D could have been made from type A, B rocks by adding 1 to 2 percent of carbonaceous-chondrite-like material. Opik predicted a meteoritic component of 3 percent of unspecified composition (*2a*).

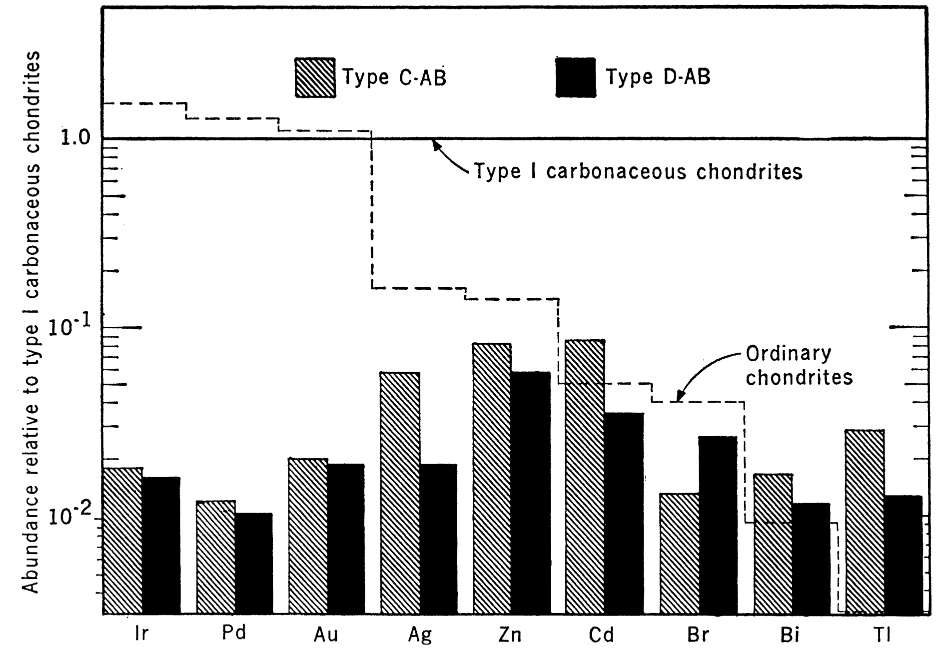

Fig. 1. Compared to A, B rocks, breccia (C) and soil (D) are enriched in nine trace elements. "Excess" component, obtained by subtracting average A, B abundances from C and D, shows abundance pattern resembling that of carbonaceous chondrites, not of ordinary chondrites. Apparently the enrichment was caused by the addition of 1 to 2 percent carbonaceous chondrite-like material.

The match is not perfect: Zn, Cd, and one of the Ag points fall higher, at 3 to 8 percent. Perhaps our A, B "average" is unrepresentative, failing to include some Zn, Cd-rich rock. Indeed, data on major elements, lanthanides, and Hf (*3, 4*) suggest that a straight A, B average does not quite match C, D composition; some particular B rocks (for example 44 and 45) perhaps having contributed to a disproportionate degree (*4*). Alternatively, the "extraneous" component may have been richer in Zn, Cd than are type I carbonaceous chondrites. Figure 1 sets a few limits on the composition of the extraneous component. Type I and II carbonaceous chondrites and type I enstatite chondrites are acceptable source materials, as are other types of relatively undifferentiated cosmic matter: solar-wind and cometary material. (The high water content of some of these materials does not disqualify them, as such water would evaporate during impact and quickly be lost from the moon.) Ordinary chondrites and iron meteorites are excluded as major contributors: the former contain far too little Ag, Br, Bi, and Tl; and the latter, too much Ir and Au in relation to the remaining elements.

The solar wind, too, seems to have made only a minor contribution. The present-day proton flux is 2×10^8 cm^{-2} sec^{-1} (*5*); assuming a cosmic Ir/H ratio of 1.65×10^{-11} (*6*), we obtain an integrated flux of 4.9×10^{-7} g Ir/cm^2 in 4.5×10^9 years. The average thickness

Table 1. Abundances in Apollo 11 lunar samples and terrestrial rocks. Doubtful values are italicized. BCR-1 = Columbia River basalt; Knippa = Knippa basalt; APG = titanium-rich Adirondack pyroxene gabbro, 1 mile SSW of Brown Point, Willsboro Quadrangle. We are indebted to A. F. Buddington for this sample. Powdered material is available from G. G. Goles, Department of Geology, University of Oregon.

Sample	Type	Ir (ppb)	Au (ppb)	Zn (ppm)	Cd (ppb)	Ag (ppb)	Bi (ppb)	Tl (ppb)	Br (ppb)	Pd (ppb)	Co (ppm)	Cu (ppm)	Ga (ppm)	Rb (ppm)	Cs (ppb)	In (ppb)
20,25	A	0.027	0.075	1.29	6.37	2.27	0.15	0.33	19.2	1.5	5.65	6.57	1.90	0.74	30.6	*15*
57,31	A	0.023	0.017	1.71	3.15	0.69	0.27	1.09	21.7	7.3	27.2	3.52	> 3.5	3.68	159	*3*
72,23	A	*4.02*	0.14	1.81	6.47	*17.3*	0.73	0.92	76	3	27.2	4.94	4.73	5.98	159	*179*
50,26	B	0.007	0.031	1.75	2.56	1.42	≦0.16	0.33	7.6	1.4	15.2	15.2	4.41	0.60	25.9	*4*
47,32*	B	*0.240*	*0.33*	*5.76*	*255*	*24.7*	*2.15*	0.57	*133*	2	14.4	13.3	5.35	1.25	44.6	*109*
48,31	C	6.88	2.66	28.6	78.3	23.6	1.62	2.83	119	13.0	34.8	9.19	5.85	4.15	128	*96*
61,31	C	9.18	3.42	29.2	106	*163*	2.79	2.70	69.2	7	34.2	22.0	5.79	3.99	146	*1430*
84,49	D	7.62	4.15	21.1	53.3	8.89	1.55	1.65	126	11.0	26.8	8.07	5.41	3.33	98	*524*
84,49	D	6.93	2.01	21.0	35.4	8.60	1.37	1.47	159	9.4	28.1	7.75	5.24	3.09	94	*768*
84,49W†	D	6.88	2.38	19.5	29.6	8.67	1.97	3.54	187	8.9	27.0	8.64	5.25	3.22	96	*1470*
84,49,1†	D	6.28	2.87	10.6	28.3	4.33	1.43	7.35	139	8.3	26.1	6.99	4.89	3.36	99	*12*
84,49,2†	D	6.67	1.99	21.0	28.3	8.53	1.96	1.91	189	9.2	26.7	8.94	5.34	3.17	94	*1950*
84,49,3†	D	10.77	4.41	34.2	46.0	24.1	3.77	6.74	317	8.6	32.5	11.2	5.72	3.27	104	*1690*
BCR-1	Bas.		0.43	127	99.0	26.4	40.2	274	97	13.0	35.4	17.1	20.1	44	930	96
BCR-1	Bas.		1.33	126		26.7	44.4	283	152	11.0	36.7	18.8	22.5	48	920	107
Knippa	Bas.		2.85	125			88.3	20.3	1300		76.8	15.0	20.2	26	745	115
APG	Gab.		3.59	125	160	34.8	5.84	24.5	73.8		67.6	17.6	1.41	3.31	104	112
A, B ave.		0.066	0.041	1.64	4.64	1.46	0.33	0.63	31.1	3.4	18.8	7.54	3.68	2.75	99	≦4
C ave.		8.03	3.04	28.9	92	23.6	2.20	2.76	94	9.9	34.5	15.6	5.82	4.07	137	≦100
D ave.		7.14	2.85	20.5	39.4	8.72	1.63	2.22	157	9.1	27.3	8.15	5.30	3.21	96	≦500

*Powder prepared at LRL; probably contaminated. †Sieve fractions: 1 = +100 mesh (24%); 2 = 100 to 325 mesh (69%); 3 = −325 mesh (7%); W = weighted average of individual analyses of fractions 1, 2, 3.

of the lunar regolith in the southern Sea of Tranquillity is 4.6 m (*7*). For a mean packed density of 1.8 g/cm^3, a 10 percent content of large rock fragments, and complete mixing, one would thus expect a mean Ir content of 2.0×10^{-10} grams of Ir/g, only about 2.7 percent of the observed value in our C and D samples. Perhaps the solar wind flux was higher in the past, but it is interesting that the observed Xe content in lunar soil, 3.6×10^{-7} cm^3 per gram at standard temperature and pressure (*8*) agrees with the amount expected for the present-day flux, 3.9×10^{-7} cm^3 per gram at standard temperature and pressure. The Ir content of C and D material can be used, though, to set an upper limit of $\leq 8 \times 10^9$ cm^{-2} sec^{-1} to the mean solar wind proton flux during the past 4.5 eons.

It appears that the bulk of the extraneous component is meteoritic or cometary. With the same assumptions as before, we find that an influx of 2.9×10^{-9} g cm^{-2} yr^{-4} of carbonaceous–chondrite-like material is needed to account for the observed average excess of Ag, Au, Bi, Br, Ir, Pd, and Tl. This figure agrees fairly well with an analogous estimate for the earth, $(1.2 \pm 0.6) \times 10^{-8}$ g cm^{-2} yr^{-1}, obtained by Barker and Anders (*9*) from the Ir and Os content of Pacific sediments. The agreement is even better if one allows for the disproportionately greater gravitational enhancement of the earth's capture cross section.

Not too much should be made of this agreement, though. The terrestrial value is an average for the last 10^5 years over some 10^7 to 10^8 km^2, while the lunar value is an average for $>10^9$ years at a single location. Both the mechanisms and efficiencies of lateral and vertical mixing are surely different for the two bodies, particularly for large projectiles that contribute most of the incoming mass. The compositional difference between C and D material, and the presence of a stratigraphic boundary in one of the Apollo 11 core samples (*8*) show that the Tranquillity Base regolith is not well mixed; thus our results for C and D samples may not be representative of the regolith as a whole. The apparent predominance of carbonaceous chondrite material is of interest, however. Several authors (*2, 10*) have previously suggested that carbonaceous chondrites comprise the major part of the meteoritic influx, but are underrepresented in collections because their friability leads to preferential destruction in the atmosphere.

Table 2. Thorium and uranium abundances in these lunar samples.

	^{232}Th (ppm)	^{238}U (ppm)	Sample
Chicago	2.0 ± 0.2	0.70 ± 0.8	72,41
Houston	2.9 ± 0.4	0.75 ± 0.11	
Chicago	2.5 ± 0.2	0.38 ± 0.15	18,24
Houston	2.3 ± 0.3	0.60 ± 0.09	
Chicago	2.2 ± 0.2	0.73 ± 0.06	61,21

The trace element enrichment in the − 325-mesh fraction of the fines (84, 49,3) is consistent with either an impact or a solar wind origin. Material closest to the impact center will be comminuted most severely and will receive the greatest admixture of projectile matter. Grain crushing by subsequent impacts would be an additional mechanism tending to enrich the finest fraction in meteorite material.

Some highly tentative conclusions about the moon may be drawn from a comparison of A, B rocks with other materials. A suitable terrestrial reference material is the Adirondack pyroxene gabbro (APG) (Table 1) whose mineralogy and titanium content match that of the Apollo 11 rocks (*11*). Of the elements analyzed by us, Ag, Au, Cd, Zn, In, Tl, and Bi are 1 or 2 orders of magnitude less abundant in A, B rocks than in the pyroxene gabbro, while most others, including some major elements measured by Schmitt (private communication), agree within a factor of 3. Essentially similar trends are found if average continental basalts (*12*) are used for comparison.

It is surprising that siderophile elements (Co, Ir, Au) are more depleted in A, B lunar rocks than in their terrestrial analogs. Apparently the removal of siderophiles from the crust was more efficient on the moon, although its metal-phase content is at most 1/10 that of the earth. Perhaps conditions were more reducing, or the scarcity of water and sulfur depressed the solubility of siderophiles in the silicate phase.

The remaining elements (Ag, Cd, Zn, In, Tl, and Bi) are volatile and have large ionic radii; they are therefore expected to concentrate rather strongly in the crust (*13, 14*). The most straightforward inference to be drawn from their low abundance is that the moon accreted at higher temperatures than did the earth (that is, above 600° to 700°K), and acquired a lesser proportion of low-temperature volatile-rich material in the terminal stages of accretion: perhaps only 10^{-4} to 10^{-5} lunar masses. The scarcity of water and carbon certainly agrees with this view. But the alternative, that the volatiles were first accreted and then lost in a high-temperature stage, cannot be completely excluded. Loss by exospheric escape requires unreasonably high temperatures, around 3000°K, for elements such as Bi and Tl. But loss by vaporization into a low molecular weight atmosphere which is subsequently swept away requires only about 1500°K. Although this mechanism requires rather special circumstances, one cannot yet rule it out altogether. We must also bear in mind the possibility that the major part of the moon's Bi, Tl, and so on is located elsewhere in the lunar crust, or retained in the interior to a greater degree than on earth.

Our data show numerous interelement correlations in A, B rocks, only one of which will be mentioned here. The covariance of Tl with Rb and Cs suggests that Tl is mainly lithophile in the lunar crust, as in the earth but not in the chondrites.

In a separate set of experiments carried out by authors Herzog and Jeffery, the positron activities of five small lunar samples (9 to 29 g) were measured by gamma-gamma coincidence spectrometry. One of the spectrometers has been described previously (*15*); the other, with 7.6 cm × 3.8 cm and 7.6 cm × 10.2 cm NaI detectors, had circuitry for two-parameter analysis and was operated at the bottom of a 16-m well for maximum background reduction.

Counting efficiencies for ^{22}Na, ^{26}Al, ^{44}Ti, ^{46}Sc, ^{56}Co, ^{58}Co, ^{60}Co, ^{232}Th, and ^{238}U were determined by means of individual mock-ups (*15, 16*) containing known amounts of the radionuclides dispersed in a matrix of Fe, MgO, or dunite powder matching the lunar sample in bulk density.

In order to determine total positron activities, gross counting rates in the 511-511 kev coincidence channel were corrected for contributions from ^{232}Th and ^{238}U, typically about 5 percent. All but 5 percent of the positron activity was due to ^{22}Na and ^{26}Al. Rock 18,24 exhibited a positron activity of 107 ± 10 β^+ min^{-1} kg^{-1} in good agreement with the preliminary result (*8*) of 135 ± 25 β^+ min^{-1} kg^{-1} despite the fact that our sample was only a 25.6 g fragment of the 213 g rock counted by the preliminary examination team. Our result for sample 72,41, of 190 ± 20 β^+ min^{-1} kg^{-1}, on the other hand, lies nearly a factor of two above that found for the whole rock (*8*) (97 ± 18 β^+

min^{-1} kg^{-1}). Since variations in composition are presumably too small to account for such a large difference we infer that sample 72,41 came from near the top surface of the rock where it was exposed to the low energy component of solar and galactic cosmic radiation. On the same basis we deduce that 26,10 (an entire small rock with an activity of 217 ± 20 β^+ min^{-1} kg^{-1}) was also exposed while 44,36 (an 11.2 g fragment of a 248 g rock with an activity of 88 ± 7 β^+ min^{-1} kg^{-1}) was shielded. The type C rock fragment 61,21 had a positron activity of 103 ± 5 β^+ min^{-1} kg^{-1} (*17*).

While the positron activities of these small samples can be determined to within 5 percent in a week's counting, accurate resolution of ^{22}Na and ^{26}Al requires counting times of a month or more. We hope to present data for ^{22}Na and ^{26}Al at a later time. In contrast, Th and U can typically be determined to within better than 20 percent in a week's counting time; data for three samples are given in Table 2. The results are generally in accord with those of the preliminary examination team (*8*).

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17. Positron activities quoted have been corrected to 12/1/69.
18. We are indebted to A. Pierce and F. Quinn for technical assistance, and to M. E. Lipshutz of Purdue University for the use of counting equipment. We gratefully acknowledge the assistance of G. F. Herman in counting the samples, and of J. R. Naples in programming and data reduction. This project was supported by AEC contract AT(11-1)-382 and NASA contract NAS 9-7887.

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Potassium, Rubidium, Strontium, Barium, and Rare-Earth Concentrations in Lunar Rocks and Separated Phases

Abstract. *Concentrations of potassium, rubidium, strontium, barium, and rare-earth elements have been determined by mass spectrometric isotope dilution for eight Apollo 11 lunar samples and for some separated phases. Potassium and rubidium are at chondritic levels, strontium at 15 times, and barium and rare earths at 30 to 100 times chondritic levels. There are trace element similarities between the lunar samples and basaltic achondrites, terrestrial dredge basalts and the bulk earth. The trace element data appear to be consistent with these lunar samples being the result of limited partial fusion of some material similar to the brecciated eucrite meteorites.*

Concentrations of K, Rb, Sr, Ba, and rare-earth elements in eight Apollo 11 lunar samples, in a fine fraction from the soil, and in mineral fractions separated from two of the igneous rocks have been determined. Preliminary values for K, Rb, Sr, Ba, and Yb contents in Apollo 11 samples, obtained by optical spectrography and gamma-ray spectrometry, have been presented (*1*). Our trace-element analyses were done by mass-spectrometric stable-isotope dilution (*2*). The precision of our whole-rock data is, with a few random exceptions, better than about ±2 percent; accuracy is believed to be quite similar. The quality of the mineral separate data is somewhat poorer because of significant blank corrections, which exceed 10 percent in three cases, and because of less than optimum spiking. Our analytical data are given in Table 1. Values for four of the whole rocks, normalized to average chondritic meteorite concentrations (given in Table 1), and covering the observed concentration range of our samples, are shown in Fig. 1. The order in which the elements are presented in the table and figures was selected to permit ready estimation of important elemental ratios.

It is apparent from Table 1 and from Fig. 1 that to a first approximation, all our whole-rock samples are quite similar in trace-element characteristics. It may be significant that two of our three igneous rocks are nearly identical in all of the trace elements studied, and that the four breccias are very similar to each other; a limited number of provenance sites may be indicated for the Apollo 11 samples.

The relationships between the trace element concentrations in the lunar samples and in the average chondritic meteorite are apparent in Fig. 1. The lunar samples have trace element characteristics similar to those of the basaltic achondrite meteorites (*3, 4*). The similarity is brought out in Fig. 1 by our data for Juvinas, a typical brecciated eucrite. There is some difference in the absolute level of the trace element concentrations. The level in Stannern is the highest that we know of in the basaltic achondrites, being about double that of Juvinas; Stannern is also of interest in showing a definite negative Eu anomaly. The similarity in K/Ba for the lunar samples and the basaltic achondrites may be particularly significant, inasmuch as this ratio tends not to change much during igneous differentiation (*5*). Angra dos Reis, a unique titaniferous-augite achondrite, has rare-earth concentrations (*4*), including a negative Eu anomaly, that are distinctly similar to those in the lunar samples. However, Ba is lower, and K and Rb much lower, in Angra dos Reis. It might be significant that sample 62 displays a tendency in this direction relative to the other lunar samples; sample 62 may have contained considerable cumulative feldspar and pyroxene at some stage in its evolution.

Trace-element concentrations in the terrestrial continental crust (*6*) are quite different from those in the lunar samples. K and Rb are about an order of magnitude higher, Sr about three times higher, and the heavier rare earths a factor of 3 to 4 lower than in the lunar samples. Tektite trace element contents resemble those of the continental crust rather than those of the lunar samples. The lunar samples show a marked resemblance to later differentiates dredged from the ocean floor (*7, 8*). The similarity might arise from their both possibly represent-