Reports

Volatile Elements in Apollo 16 Samples: Possible Evidence for Outgassing of the Moon

Abstract. *Several Apollo 16 breccias, including one containing goethite, are strikingly enriched in volatile elements such as bromine, cadmium, germanium, antimony, thallium, and zinc. Similar but smaller enrichments are found in all highland soils. It appears that volcanic processes took place in the lunar highlands, involving the release of volatiles including water. The lunar thallium/uranium ratio is 2×10^{-4} of the cosmic ratio, which suggests that the moon's original water content could not have exceeded the equivalent of a layer 22 meters deep. The cataclastic anorthosites at the Apollo 16 site may represent deep ejecta from the Nectaris basin.*

We have analyzed 27 Apollo 16 samples for 17 trace elements, using neutron activation analysis. The results, reported in full elsewhere (*1*), have implications for the local geology of the Apollo 16 site as well as two topics of global scope: the moon's overall depletion in volatile elements (*2*) and the presence of an ancient meteoritic component from the final stages of the moon's accretion (*3*).

One of the most striking features of the moon is the great depletion in volatiles, such as C, N, H_2O, Pb, Bi, and Tl. Apparently these elements were largely left behind in the solar nebula when the moon accreted (*2*). For an understanding of the moon's chemistry, it would be of interest to know the magnitude of this depletion relative to cosmic or terrestrial abundances. The only elements for which this can be estimated with some confidence are Tl and Pb. Unlike C, N, and H_2O, they are not lost from the lunar surface and hence not derived mainly from the solar wind. And more so than Bi, they tend to concentrate in the crust during planetary differentiation (*2, 4, 5*). Thus, their whole-planet abundance can be estimated from their crustal abundance.

In practice, one compares the abundance of Tl or Pb with that of geochemically similar but less volatile elements (K, Rb, Cs, and U; the latter is particularly suitable because of its low volatility). All are lithophile, form large ions, and do not separate readily from each other in magmatic processes, as indicated by the near-constancy of the K/U, Pb/U, K/Rb, and Tl/Rb ratios in hundreds of rocks from a particular planet (*2, 4, 6*).

Lunar rocks from the first five landing sites show rather good Tl-Cs and Cs-U correlations over nearly three orders of magnitude in Cs content [Fig. 1a and (*2*)]. From the Tl/U ratio of 2.8×10^{-3}, it appeared that the moon was depleted in Tl by a factor of 2.3×10^{-4} relative to cosmic abundances, compared to a factor of 2.7×10^{-2} for the earth (*2*). However, with the return of highland rocks by Apollo 15, Apollo 16, and Luna 20, it has become apparent that lunar anorthosites (rocks containing more than 60 percent plagioclase feldspar) often deviate strikingly from this trend (Fig. 1b). Most anorthosites from Apollo 15 have too little Tl in relation to their Cs or U content, and most anorthosites from Apollo 16, too much.

Why do the anorthosites fall out of line? Crystal chemistry cannot be the answer, because if the lattice can hold Cs^+ (radius 1.78 Å) it should be able to hold Tl^+ (radius 1.58 Å). Brecciation likewise does not seem to be the key factor, because the great majority of some 60-odd breccia particles from soils are not notably enriched in Tl (*1*). More likely, the anomaly is due to the volatility of Tl. At least four other volatile elements (Br, Cd, Sb, and Ge) correlate with Tl, showing the highest abundances in the rocks richest in Tl (Table 1). Presumably all were fractionated together.

Table 1. Trace elements in Apollo 16 samples. Concentrations are in parts per billion, except for Zn and Rb, which are given in parts per million. Complete data, including 11 more samples and three elements (Ag, Ni, and Se) will be published elsewhere (*1*). Abbreviation Cl means classification. Rocks are classified by composition and nature of dominant mafic mineral: A, anorthosite; AG, anorthositic gabbro; AN, anorthositic norite; AT, anorthositic troctolite; S, soil (< 1 mm). The numbers give the classification according to the Apollo 16 Preliminary Examination Team (*13, 14*): 2, cataclastic anorthosites; 3, igneous and high-grade metamorphic rocks. The first four soils are from the South Ray Crater area, the last four, from the vicinity of North Ray Crater.

Sample	Cl	Ir	Re	Au	Sb	Ge	Te	Br	Bi	Zn	Cd	Tl	Cs	Rb	U
60025,84	AT2	0.0057	0.0016	0.0074	0.035	2.30	65	265	3.58	0.17	7.25	26	2.0	0.017	0.92
61016,156	A2	0.0099	0.0022	0.020	0.15	13	≤0.4	350	0.22	1.45	190	131.2	1.2	0.018	0.88
61916,132	AG	11.5	1.28	9.55	3.13	620	4.8	850	7.3	0.84	29.5	45.3	84	2.0	515
61016,132	AG	6.67	0.841	5.60	1.74	353	5.7	410	2.9	0.74	9.6	22.7	56	1.3	485
62295,40	AN3	3.58	0.336	3.10	0.88	642	12.5	50	0.26	16.5	4.9	4.29	530	5.8	935
65015,51	AN3	11.6	1.16	10.2	4.34	380	3.25	22	0.099	0.48	9.25	0.22	405	7.8	3420
66095,55	AG3	16.6	2.13	17.9	6.9	2140	20	826	0.27	50.5	328	175	160	3.9	1020
68415,67	AG3	4.58	0.434	2.65	0.53	73	4.7	140	0.052	1.45	0.85	4.6	80	1.4	445
61221,6	S	6.21	0.578	4.94	1.60	345	14.7	165	1.10	21.5	140	7.4	90	2.0	431
61241,6	S	9.64	0.762	5.31	1.55	582	15.6	160	0.91	22.5	119	6.6	105	2.2	538
64501,9	S	11.8	0.996	13.8	2.20	485	22.8	235	1.06	23.8	85.5	7.9	85	2.0	580
68841,15	S	12.8	1.56	9.02	2.45	875	21	130	0.88	18.7	56.5	4.80	130	3.0	670
63321,1	S	8.29	0.669	8.60	1.23	404	17.3	145	0.72	17.0	78	4.1	83	1.8	401
63341,1	S	11.10	0.741	7.08	1.51	400	18.5	125	0.76	15.5	57.5	4.0	86	1.9	398
63501,13	S	11.21	0.955	8.51	1.85	655	13.6	85	0.63	12.0	53.5	2.8	72	1.6	382
67601,19	S	5.01	0.527	2.47	0.73	245	8.5	80	0.32	6.9	21	0.98	54	1.3	295

It is unlikely that these elements were injected from a volatile-rich projectile, such as a comet nucleus (*7, 8*). Judging from the micrometeorite component in lunar soils, comets seem to have a carbonaceous–chondrite-like (or C1) composition (*2*). But the volatiles in Apollo 16 breccias do not occur in C1 chondrite proportions (for example, Cd/Zn in 61016,156 is about 60 times the C1 ratio), and the enrichments are too high (Tl is present in up to 120 percent the C1 value). Moreover, the Tl-enriched breccia 66095 shows a parallel enrichment in Pb, of decidedly lunar, not meteoritic, isotopic composition (*9*).

It also seems improbable that the enrichment took place by vaporization in a hot ejecta blanket, as proposed by McKay *et al.* (*10*) for Apollo 14 breccias. None of the Apollo 14 breccias studied by us (*2*) show abundance anomalies for the volatile elements in question.

We suspect that the fractionation took place by volcanic activity (fumaroles?) after formation of the rocks, one of the mechanisms considered by Agrell *et al.* (*11*), Williams and Gibson (*12*), and McKay *et al.* (*10*). Some evidence for alteration by H_2O was obtained by the Apollo 16 Preliminary Examination Team (*13, 14*) who observed goethite (FeOOH) rims around metal grains in breccia 66095, similar to but far more abundant than those seen by Agrell *et al.* (*11*) in Apollo 14 breccia 14301. Indeed, 66095 shows the greatest enrichment in Sb, Ge, Zn, Cd, Tl, Se (Table 1), and Pb (*9*), and even contains detectable zinc minerals (*8*). These elements, along with As, Hg, W, and others, often are enriched in terrestrial hot springs and fumaroles (*15*). That this process happened after crystallization of the rocks is suggested by the similarity in Tl content of both the light and dark portions of 61016 (Table 1), in spite of their marked compositional differences. Nunes *et al.* (*9*) have inferred a date of $(3.77 \pm 0.02) \times 10^9$ years for this process in 66095, from the isotopic composition of the Pb.

Table 2. Thallium/cesium ratio in lunar soils.

Site	Tl/Cs
Highland	
Apollo 14	3.6×10^{-2}
Apollo 15	2.3×10^{-2}
Apollo 16	5.4×10^{-2}
Luna 20	5.6×10^{-2}
Average	4.2×10^{-2}
Mare	
Apollo 11	1.6×10^{-2}
Apollo 12	0.9×10^{-2}
Apollo 15	1.2×10^{-2}
Luna 16	2.2×10^{-2}
Average	1.5×10^{-2}

It would seem that this enrichment process, whatever its nature, is not restricted to the Apollo 16 site. Highland soils in general seem to be enriched in Tl, judging from the fact that their Tl/Cs ratios fall consistently above the average ratio for rocks, 1.0×10^{-2} (Table 2). Mare soils, on the other hand, show no significant enrichment when a small meteoritic contribution is taken into account. Similar enrichments in soils relative to rocks have been seen for Zn, Cd (*2, 3*), and Pb (*5*), especially in the highlands. Perhaps we are seeing the effects of an early volcanism, more pronounced in the highlands because of their age or chemistry.

The new data for the highlands force a reappraisal of the moon's overall abundance of volatiles. A fairly good value for the uppermost highlands crust can be obtained from the soils, which are representative of a wider geographic area than the rocks. The average Tl/U ratio for the highland soils is 8.8×10^{-3}, compared to 2.3×10^{-3} for highland and mare rocks other than anorthosites. The true value for the lunar crust probably lies close to the lower ratio, because the Tl enrichments at the surface must have been offset by complementary depletions at depth. The lunar Tl/U ratio thus is 1.9×10^{-4} of the cosmic ratio,

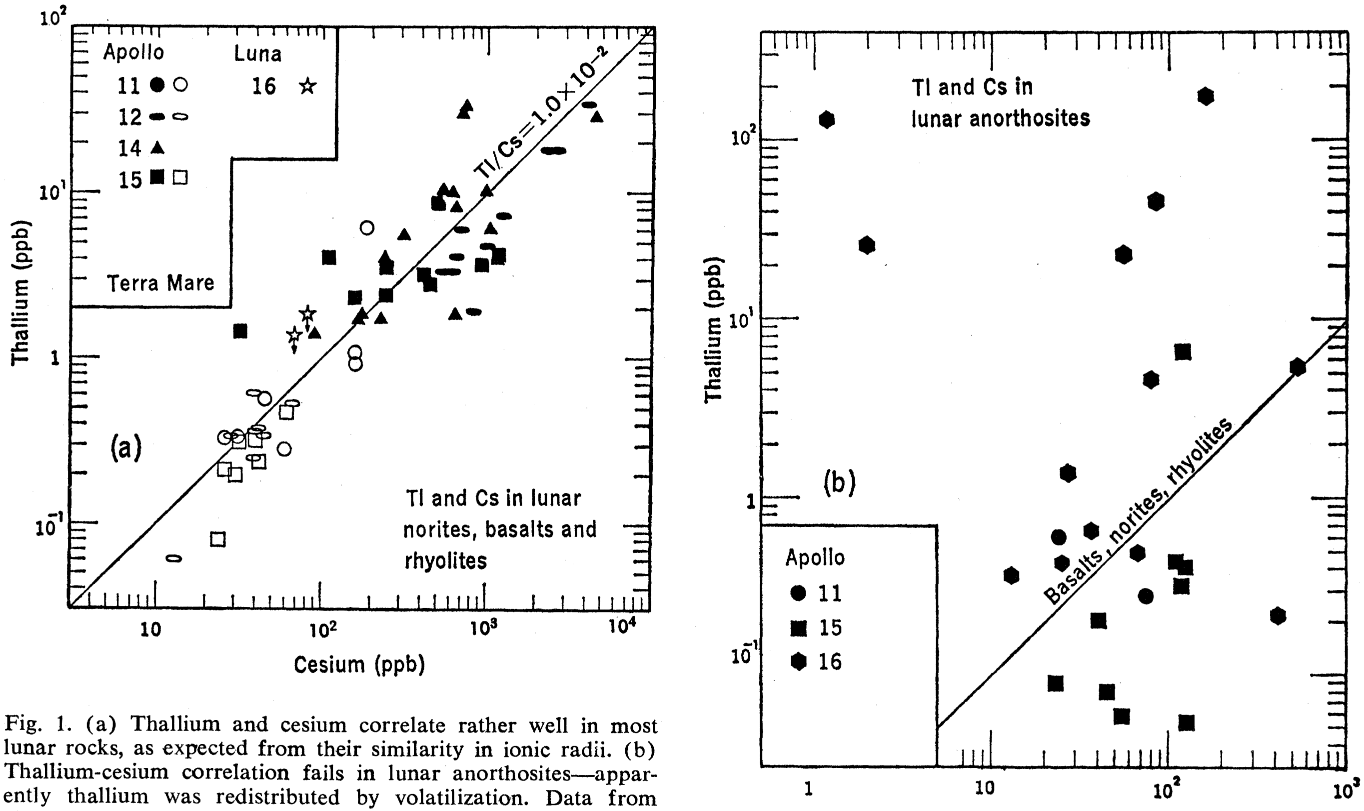

Fig. 1. (a) Thallium and cesium correlate rather well in most lunar rocks, as expected from their similarity in ionic radii. (b) Thallium-cesium correlation fails in lunar anorthosites—apparently thallium was redistributed by volatilization. Data from Table 1 and (*1, 2*).

nearly identical to the earlier estimate of 2.3×10^{-4}, and still lower by a factor of 100 than the corresponding value for the earth [2.2×10^{-2}, according to the latest data (*16*)]. The analogous ratios for Pb, from the data of Tatsumoto and co-workers (*5*), are very similar: 3.1×10^{-4} and 1.8×10^{-2}.

We can interpret these values in terms of the two-component model of meteorite formation (*2, 17*), which may be valid for planets, at least to a first approximation (*4, 17*). According to this model, planets are a mixture of low-temperature material containing all the elements condensed from the nebula up to the time of accretion (such as U, Cs, and part or all of the Tl and Pb), and high-temperature material containing only the less volatile elements (such as U but not Tl or Pb). It is very significant that Tl and Pb are present in nearly their cosmic ratio in both the earth and the moon. Since Tl is more volatile than Pb (*17*), the low-temperature material must have formed at a temperature low enough for both elements to be fully (at least 50 percent) condensed, that is, below 470°K at 10^{-3} to 10^{-4} atm (*17*). In terms of this model, the Tl/U ratios of 1.9×10^{-4} and 2.2×10^{-2} for the moon and the earth, multiplied by the enrichment factors of U, give the mass fraction of low-temperature material in the two planets.

It seems likely that the composition of the low-temperature fraction changed during accretion, in response to temperature changes in the nebula. Any material equilibrated with the gas below 380° to 400°K would contain H_2O in the form of hydrated silicates similar to those in carbonaceous chondrites. Following Anders (*18*), we can estimate an upper limit to the moon's water content, on the assumption that the earth and the moon acquired Pb, Tl, and H_2O from a volatile-rich material of cosmic Pb and Tl content and an unknown but fixed H_2O/Tl ratio. The initial water content of the moon, W_m, can then be found from the mass of the earth's hydrosphere, W_e, by a simple proportion:

$$W_m/M_m = W_e U_m (\mathrm{Tl/U})_m / M_e U_e (\mathrm{Tl/U})_e$$

where M_m and M_e are the masses of moon and the earth, and U_m and U_e are the bulk uranium contents. With $W_e = 1.45 \times 10^{24}$ g, $U_m = 90$ ppb, $U_e = 18.7$ ppb (*19*), and the above Tl/U ratios, $W_m = 1.2 \times 10^{-5}\ M_m$, corresponding to 2200 g/cm², or a layer 22 m thick. This value, which should be regarded as upper limit, corresponds to $\leq 1.2 \times 10^{-4}$ of the mass of the lunar crust, in accord with the extreme scarcity of hydrated minerals in lunar rocks.

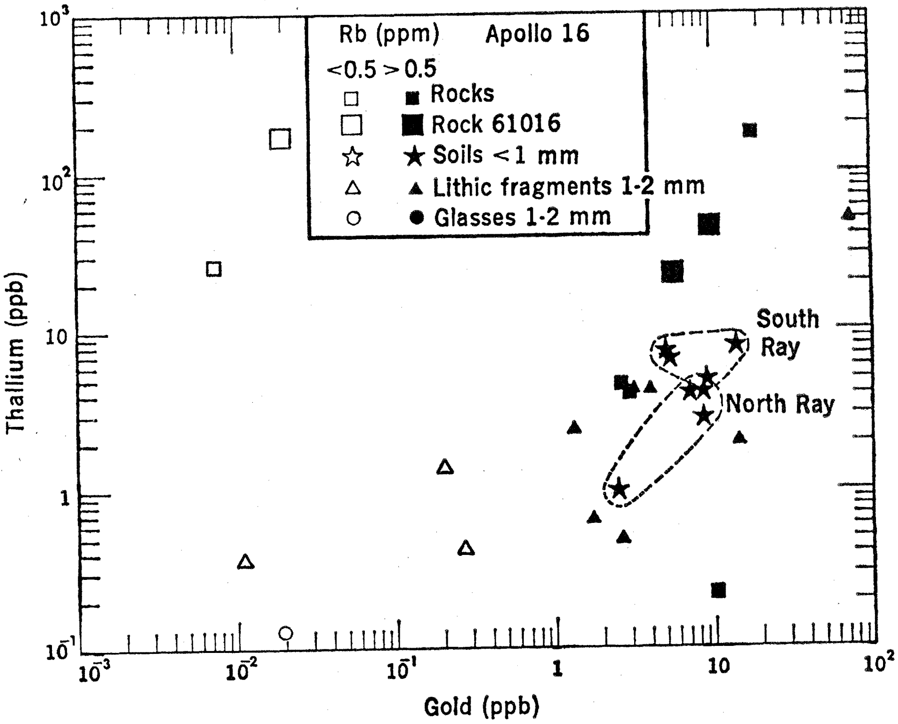

Fig. 2. Alkali-rich rocks have much higher Au contents than do alkali-poor rocks, reflecting the presence of an ancient meteoritic component. Apparently the latter come from a deeper stratum, uncontaminated by meteoritic material. High Tl contents are found in both rock types, which suggests that the Tl enrichment was not caused by meteorite infall.

The Apollo 16 samples contain the same "ancient" meteoritic component that has been found in most highland breccias (*3*). It apparently represents debris of planetesimals that bombarded the moon during the first $\sim 0.7 \times 10^9$ years of its history, and accumulated in the ancient highlands regolith. Six of the eight Apollo 16 rocks are enriched in the siderophile elements Ir, Re, and Au, which are diagnostic of meteoritic material. There is little doubt that this material represents an ancient component and not recent meteoritic contamination. The rocks in question have remained closed systems for $(3.9 \text{ to } 4.4) \times 10^9$ years [for example, 65015 (*20*)], and have low Ir/Au, Re/Au, and Bi/Au ratios which fall outside the meteoritic range but are characteristic of the dominant type of ancient component at other sites.

The siderophile elements and alkalis can serve as "markers" for determining the stratigraphy of the Apollo 16 site (Fig. 2). The alkali-rich rocks (anorthositic gabbros and norites) have Au contents greater than 2 ppb, whereas the alkali-poor rocks (cataclastic anorthosites and troctolites) have Au contents of less than 0.02 ppb. Lithic fragments and glasses picked from coarse soils (1 to 2 mm) (*1*) show the same trend, though less clearly owing to slight contamination by meteoritic material.

It is interesting to relate these rock types with the three stratigraphic units exposed by North Ray Crater (*20*). The rocks rich in Au and Rb can probably be identified with the light-matrix breccias which comprise the top 100 m, and the rocks poor in Au and Rb with the light, friable unit immediately below. A possible representative of the third unit, a dark layer on the crater floor, is rock 67915, a breccia sample from a large boulder next to House Rock on the crater rim (*13, 14, 21*). Matrix and clasts from this rock (*1*) again have fairly high contents of Au (1.90 and 1.06 ppb) and Rb (0.80 and 1.1 ppm), rather like the rocks in Table 1 tentatively assigned to the topmost unit. But they differ from these rocks in having a nearly threefold higher Re/Au ratio. The Re/Au or Ir/Au ratio is a useful criterion for distinguishing different types of ancient meteoritic component (*3*), and it thus seems that rocks from the dark bottom layer contain a distinctly different type of meteoritic material.

All three units probably represent ejecta from nearby mare basins. At the time the last mare basins were formed, the highlands surface probably was covered with a thick regolith of ejecta from older basins, containing crushed and brecciated rocks along with debris of the impacted planetesimals. The anorthosites poor in Au and Rb at Apollo 16 apparently come from deep strata well below this regolith, uncontaminated by projectile debris. The most likely source of deep ejecta is the Mare Nectaris basin, fifth in age among major frontside basins and only 2.8 basin radii from the Apollo 16 site (*22*). If this identification is accepted, then the following assignments result.

1) Dark unit: shallow ejecta from Nectaris basin.

2) Light, friable unit: deep ejecta from Nectaris basin.

3) Light-matrix breccias: shallow ejecta from Orientale, Imbrium, Crisium, and Humorum basins, 5.2 to 17 basin radii away.

This interpretation is supported by data on bulk soils. Soil 67601 from the rim of North Ray Crater is markedly lower in Au, Tl, Rb, Cs, and U than any other bulk soil in Table 1. Having been collected directly on the rim of North Ray Crater, it is likely to contain a larger amount of deep ejecta—mainly the light-colored, friable unit (*21*) which we identified with cataclastic anorthosites poor in Au, Rb, Cs, and U.

It is significant that Au correlates with siderophile Ir and Re (Table 1), but not with volatile Tl (Fig. 2). Apparently it was not affected by the (hydrothermal?) process that fractionated Tl and other volatiles, although under terrestrial conditions Au is often mobilized by hydrothermal fluids.

We conclude with a few observations on soils.

1) There is a slight suggestion in the data that the enrichment in Tl was limited in areal extent: four soils from the South Ray Crater area generally have higher Tl contents and Tl/Cs ratios than four soils from the North Ray Crater area (Fig. 2). The Tl-rich rocks also all seem to be South Ray Crater ejecta. Possibly the source of the Tl-rich material is localized in the southern part of the site.

2) In soil 61221, we see no perceptible enrichment of Tl and Bi paralleling the enrichment of H_2O, HCN, CH_4, and other compounds that prompted Gibson and Moore (*7*) to suggest a cometary projectile for North Ray Crater. This does not contradict their suggestion in any way, because at typical cometary impact velocities only about 0.1 percent of the projectile material would be expected in the ejecta; this would be undetectable against the background of indigenous Tl and Bi at the Apollo 16 site. By the same token, we wonder if the H_2O, HCN, and other compounds detected by Gibson and Moore do not come from the same indigenous (volcanic?) source as the excess Tl, Bi, Cd, and so on.

3) All soils contain unusually low amounts of Bi, an element diagnostic of micrometeorites in lunar soils. At the Apollo 16 site, in contrast to other sites, the Bi content of the rocks is quite variable, with three samples of high Tl content also showing high Bi abundances (Table 1). Disregarding the contribution of such rocks, we obtain a minimum indigenous correction of 0.18 ppb from the average of the remaining samples. With this correction, nominal micrometeorite components of 0.13 to 0.84 percent C1 chondrite equivalent are obtained; appreciably lower than the values of 1 to 1.5 percent that are characteristic of mature soils at other sites (*2*). Apparently the Apollo 16 soils consist largely of material of low surface exposure age—ejecta from North Ray, South Ray, and other young craters.

The Au content (and also the Ir and Re contents) suggests a much larger amount of meteoritic material—up to 9 percent nominal "C1 chondrite equivalent." But some 80 to 90 percent of this material comprises the ancient component, not micrometeorites.

4) There is no significant difference between two soils from the Descartes formation [64501 and 64801; the latter was reported in (*1*)] and seven soils from the Cayley formation, except for distinctly higher siderophile element abundances in the Descartes soils.

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